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Uranium Hexafluoride — Safe Handling, Processing, and Transporting

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FOREWORD

From the beginning of uranium enrichment activities over 40 years ago to the increasingly regulated business environment of the present, the concern for the safe handling of uranium hexafluoride continues to require a significant level of effort in the conversion, enrichment, and fuel fabrication industries. Although the safety record of the entire uranium fuel industry has often been cited for its excellence, there remains the opportunity for continued improvement.

The advent of recent technological progress, additional regulatory requirements, and increased scrutiny of the entire nuclear industry has brought a heightened interest in safe handling of uranium hexafluoride to public and private organizations around the world. As part of a continuing goal of the promotion of uranium hexafluoride handling safety, the United States Department of Energy, Oak Ridge Operations, and Martin Marietta Energy Systems, Inc., decided in 1987 to cosponsor this symposium.

This conference seeks to provide a forum for the exchange of information and ideas of the safety aspects and technical issues related to the handling of uranium hexafluoride. By allowing operators, engineers, scientists, managers, educators, and others to meet and share experiences of mutual concern, the conference is also intended to provide the participants with a more complete knowledge of technical and operational issues.

These proceedings contain the collected work of distinguished authors who have voluntarily offered to share their knowledge and expertise with the world community. The topics for the papers in the proceedings are widely varied and include the results of chemical, metallurgical, mechanical, thermal, and analytical investigations, as well as the developed philosophies of operational, managerial, and regulatory guidelines. These proceedings will be an excellent resource for those in the industry.

William D. Strunk
Technical Coordinator

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THE PHYSICAL AND CHEMICAL PROPERTIES OF URANIUM HEXAFLUORIDE*

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ABSTRACT

This paper describes what uranium hexafluoride (UF_6) is, gives some of its pertinent physical properties, illustrates significant reactions between UF_6 and other substances, touches on its toxic properties, and states some of the "do's" and "don't's" of UF_6 handling. At room temperature, UF_6 is a colorless, high molecular weight, subliming solid with a significant vapor pressure. The triple point is at 64.02°C (337.17 K) and 1.497 atm (0.1517 MPa). Because the pressure of liquid UF_6 is always above 1 atm, the behavior of a ruptured cylinder containing liquid UF_6 will be similar to that of a superheated hot water heater, but somewhat less violent. In both the solid and liquid states, UF_6 is highly expanded; that is, the number of molecules per unit volume is smaller than for most other substances. The change in density between the liquid and solid states is about one-third, an abnormally large increase.

The value of $(\partial P/\partial T)_V$ for liquid UF_6 is $4.8\text{ atm}/^\circ\text{C}$ (0.485 MPa/K) at 150°C (423.2 K). The corresponding value for solid UF_6 is $30.2\text{ atm}/^\circ\text{C}$ (3.06 MPa/K) at -40°F (233.2 K). These values help in understanding the rupture of overfilled cylinders and the bulging of cold traps. Values for other physical properties which aid in understanding the nature of the UF_6 molecule are also given.

The key to much UF_6 chemistry is the great stability of the uranyl ion (UO_2^{++}), which permits the reaction with water, oxides, hydroxides, and salts containing oxygen-bearing anions without having to liberate molecular O_2 , a high potential barrier process. The UF_6 is a relatively mild fluorinating agent but is reactive toward metals and most organic materials. Liquid UF_6 reacts with hydrocarbons with explosive violence. Silicones are destroyed by UF_6 . The UF_6 is toxic per se and is also toxic because of the HF generated by hydrolysis. The biological half-life is short because the uranyl ion is rapidly eliminated from the body by the kidneys. In closing, the implications of the properties of UF_6 are summarized in terms of a few rules for handling.

NOMENCLATURE

$(\partial P/\partial T)_V$	Rate of pressure rise per unit increase in temperature at constant liquid volume (atm/K)
P	pressure due to UF_6 acting on the cylinder walls and the pressure exerted by or upon the liquid or solid UF_6 (atm or other consistent unit)
T	temperature (K)
(dT_m/dP)	change in the temperature of fusion (melting) per unit change in pressure on the solid (K/atm)
T_m	temperature of fusion (K)
ΔV	molar volume change on melting (L)
ΔH_f	molar enthalpy of fusion (L-atm/mol)
$\rho(s)$	density of the solid (g/mL or kg/L) at temperature t
$\rho(l)$	density of the liquid (g/mL or kg/L) at temperature t
t	temperature ($^\circ\text{C}$)
t_c	critical temperature ($^\circ\text{C}$)
α	volume coefficient of expansion defined by the relation $\alpha = (1/V)(\partial V/\partial T)_P$ (reciprocal degrees)
β	coefficient of compressibility defined by the relation $\beta = -(1/V)(\partial V/\partial P)_T$ (atm $^{-1}$)
$(\partial P/\partial T)_V(s)$	rate of pressure increase per unit temperature increase in restrained solid UF_6 (atm/K)
P_{\max}	maximum pressure that could be developed in restrained solid UF_6 trapped at a given desublimation temperature and heated to another temperature differing by ΔT (K)
C_p	molar heat capacity at constant pressure (cal/mol-K), the phase to which it applies will be indicated in parentheses following [e.g., $C_p(g)$ for the gas phase]

*Based on work performed at Oak Ridge Gaseous Diffusion Plant, operated for the U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

INTRODUCTION

This paper is intended to provide a description of what UF₆ is, some of its pertinent physical properties, illustrations of the reactions between UF₆ and other substances, some concepts about UF₆ reactions important to processing, and some "do's" and "don'ts" of UF₆ handling and transporting.

At room temperature, UF₆ is a colorless, high molecular weight, nonpolar, subliming solid with significant, but less than atmospheric, vapor pressure. This statement immediately indicates that one will not be handed a bag containing UF₆ or a bottle of liquid UF₆ but that one will probably receive the UF₆ in a metal tube sealed by a valve or valves. The contents of the tube will not be subject to visual inspection and therefore must be determined by analysis.

PHYSICAL PROPERTIES

Phase Diagram¹

The phase diagram for pure UF₆ is shown in Figure 1 in which the logarithm of the vapor pressure is given as a function of the temperature. Only vapor exists in the region to the right and below the continuous curve. The liquid exists to the right of the dotted line and above the continuous line but to the left of the critical-point temperature, above which temperature the liquid and vapor are indistinguishable. The liquid range is relatively long, and the critical pressure is relatively large, about 45.5 atm (4.61 MPa), for a material of this class. As will be emphasized by others, UF₆ storage cylinders are not designed to withstand such pressures. The area to the left of the dashed and continuous curves represents conditions under which the solid UF₆ exists.

Note that the sublimation temperature is below the triple point. This has implications for processing because the

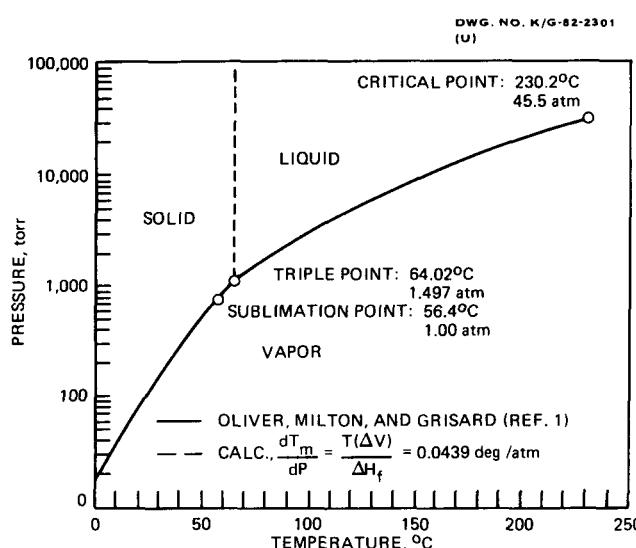


Figure 1. UF₆ phase diagram.

pressure must be above 1.5 atm (0.152 MPa) and the temperature above 64°C (337 K) for UF₆ to be handled as a liquid. Thus, any process using liquid UF₆ will be subject to leakage of the UF₆ to the atmosphere through any holes. Also, because the pressure of liquid UF₆ is always above 1 atm, the behavior of a ruptured cylinder may be similar to that of a superheated hot water heater, although somewhat less violent. Transfers below 1.5 atm or below 64°C involve moving vapor that is produced by sublimation and removed by desublimation in a cooled trap.

Density of UF₆

The UF₆ is a relatively expanded liquid and solid; that is, the number of molecules per unit volume of liquid and solid are relatively fewer than in most other materials. Still the densities are quite large, as seen from Figure 2 in which the densities of solid and liquid UF₆ are shown as functions of temperature. Equations expressing the density of the solid and liquid as a function of temperature are given below. Equation 1 comes from refs. 2 and 3, Equation 2 from ref. 4, and Equation 2a from ref. 5.

$$\rho(s) = 5.194 - 0.005168t, \text{ g/mL,} \quad (1)$$

$$\rho(l) = 1.670 + 0.15203(230.2 - t)^{0.5}, \text{ g/mL,} \quad (2)$$

or

$$\rho(l) = 2.0843 - 0.0031t + 0.3710(230.2 - t)^{0.3045}, \text{ g/mL,} \quad (2a)$$

Equation 2 is probably more accurate near the triple point, and Equation 2a is more accurate near the critical point.

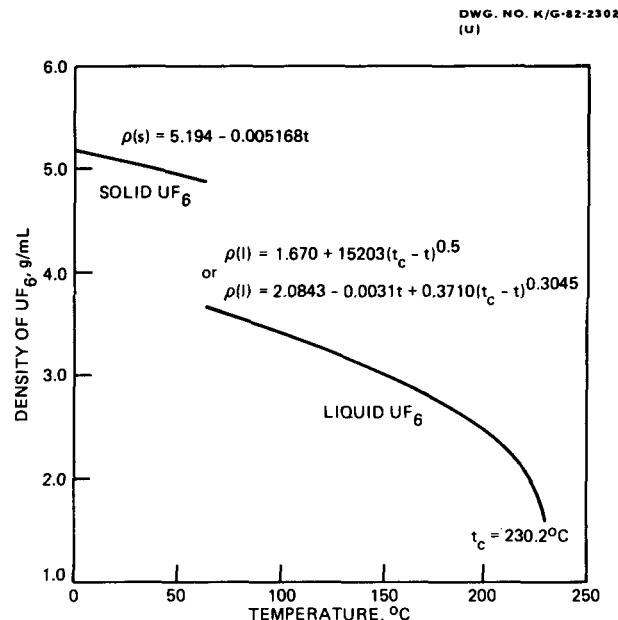


Figure 2. Density of UF₆.

Two factors influencing handling should be stressed. First, the coefficients of expansion and compressibility are noted to be relatively large for both the solid and the liquid. Rapid heating of desublimed solid may lead to trap bulging with metals and to breakage with glass containers, whether or not the trap is overfilled.³ Second, for handling liquid UF₆, sufficient freeboard (ullage) must be maintained to provide for liquid expansion for the temperature range over which the liquid is to be heated.⁴ Particular attention must be paid to the fill limits of containers when the UF₆ is to be desublimed as solid and is to be liquified for removal. The volume of the liquid produced on melting is about four-thirds of the volume of the solid.

The consequences of heating a cylinder once it has been completely filled with liquid UF₆ may be deduced from the data in Figure 3, which shows the rate of increase of the pressure of liquid UF₆ with temperature at constant volume as a function of the temperature at which the cylinder becomes filled.⁴ These values may be computed using Equation 3.

$$(\partial P/\partial T)_V = 11.42331 - 5.96051 \times 10^{-2} t + 1.02420 \times 10^{-4} t^2, \text{ atm}/\text{C}, \quad (3)$$

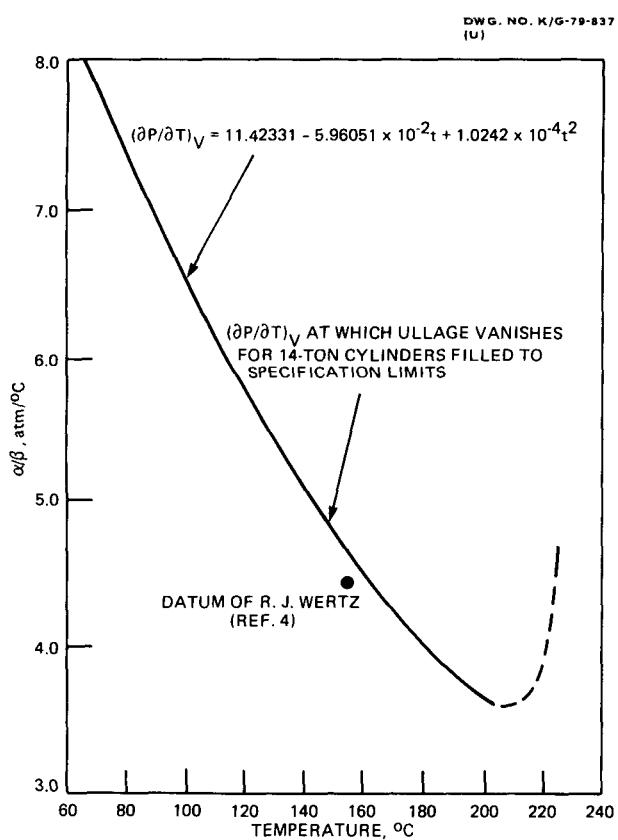


Figure 3. Rate of change of pressure with temperature at constant volume for filled UF₆ cylinders assuming an Eyring liquid.

A similar equation may be derived for use in estimating pressure in the restrained solid.³ The value of $(\partial P/\partial T)v(s)$ at a given desublimation temperature is given by

$$(\partial P/\partial T)v(s) = 2.6137 [p(s) - 2.353]^{2.06025} + 3.616, \text{ atm}/\text{C}, \quad (4)$$

so that

$$P_{\max} = (\partial P/\partial T)v(s) \times \Delta T. \quad (5)$$

The maximum pressure that could be developed by UF₆ trapped at -100°F (200 K) and heated near the melting point is 69,000 psi (476 MPa).

Other Physical Properties of UF₆

A number of properties are listed in Table 1 to give a better feel for the nature of the UF₆ molecule. Note the small value for the heat of vaporization, which means that masswise, condensation and sublimation of material may occur a lot faster than one would expect for more familiar materials with a given heat flux. The heat capacity of the vapor is large with respect to those of normal atmospheric gases, which means, for example, that UF₆ is a much more effective quenching agent than nitrogen for exothermic gaseous reactions. The

Table 1. Other Physical Properties of UF₆

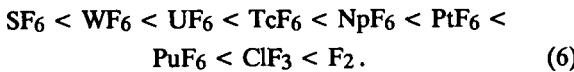
Heat of sublimation (at 64.05°C), kcal/mol	11.38
Heat of fusion (at 64.05°C), kcal/mol	4.56
Heat of vaporization (at 64.05°C), kcal/mol	6.86
Molar heat capacity of solid (at 25°C, Cp), cal/mol·°C	39.86
Molar heat capacity of liquid (at 72°C, Cp), cal/mol·°C	45.84
Molar heat capacity of vapor (at 25°C, Cp), cal/mol·°C	30.93
Cp/Cv (at 60°C)	1.067
Self-diffusion coefficient, vapor (at 30°C, 10 torr), cm ² /s	1.26
Self-diffusion coefficient, liquid (at 69.5°C), cm ² /s	1.90 × 10 ⁻⁵
Viscosity of vapor (at 25°C), micropoise	176
Viscosity of liquid (at 70°C), centipoise	0.910
Thermal conductivity of vapor (at 50°C), cal/cm·sec·°C	1.76 × 10 ⁻⁴
Thermal conductivity of liquid (at 72°C), cal/cm·sec·°C	3.83 × 10 ⁻⁴
Thermal conductivity of solid (at 50°C), cal/cm·sec·°C	1.13 × 10 ⁻⁴

combination of low surface tension, low viscosity, and high density leads one to expect quite small droplets and thus streaming in distillation columns at very low throughputs with some resultant design problems, such as how to ensure uniform wetting and flow throughout a column. Attention is called to the apparent discrepancy in the value of the triple point given in Figure 1 and Table 1 of 0.03°C. The value of 64.02°C is obtained experimentally by boiling the UF₆ under a nitrogen atmosphere. The dissolved nitrogen in the liquid UF₆ depresses the triple point by 0.03°C; thus, the triple point under the orthobaric pressure is 64.05°C.

CHEMICAL PROPERTIES

Chemistry of UF₆ (ref. 6)

The chemistry of UF₆ is largely determined by its fluorination (or oxidation) potential and the ease with which the UF₆ molecule is hydrolyzed (the U-F bond is a strongly polar bond as compared with the S-F bond in SF₆, which does not hydrolyze at all). The UF₆ is a very stable vapor having a dissociation pressure of about 10⁻³⁰ atm at 400 K in the presence of solid UF₅ (ref. 7). Thus, UF₆ is a relatively mild fluorinating agent as can be seen from its position in the series below, fluorine being the most powerful and SF₆ being inert at room temperature:



This does not mean that UF₆ does not attack metals vigorously. The UF₆ tends to be strongly chemisorbed on most materials, giving a high surface coverage and consequently good molecular contact and greatly increasing the opportunity for reaction.

Table 2 illustrates the types of chemical reaction undergone by UF₆. The key to much UF₆ chemistry is the great stability of the UO₂⁺⁺ ion, which permits reaction with water, oxides, hydroxides, and salts containing oxygen-bearing anions such as SO₄²⁻, NO₃⁻, and CO₃²⁻ without having to liberate molecular O₂, which is a high potential barrier process. Thus, UF₆ is rapidly hydrolyzed by water to UO₂F₂ or one of its solvates, depending on the quantity of water relative to the stoichiometric requirement. Metathesis reactions are illustrated by the reaction with NiO and Ni(OH)₂. The UF₆ may form adducts as illustrated by the reaction with the alkali metal fluoride, NaF, to form NaUF₇ and Na₂UF₈. The UF₆ is an oxidizing agent toward metals and is reduced to form solid products such as UF₅, U₂F₉, U₄F₁₇ and UF₄. The UF_x product produced depends on the partial pressure of the UF₆, the metal attacked, and the temperature so that the stoichiometry of the reaction is not unique.

The problem of chemisorption has already been mentioned; it occurs with almost every material except for a few fluorocarbon materials like polytetrafluoroethylene

Table 2. Chemical Properties of UF₆

- Is easily hydrolyzed as gas, liquid, or solid
 - UF₆ + 2H₂O → UO₂F₂(s) + 4HF(g)
- Carries out metathesis reactions with oxides, hydroxides
 - UF₆ + 2NiO → UO₂F₂(s) + 2NiF₂(s)
 - UF₆ + Ni(OH)₂ → UO₂F₂(s) + NiF₂(s) + 2HF(s)
- Forms "addition" complexes
 - UF₆ + 2NaF ⇌ Na₂UF₈
- Oxidizes metals and is itself reduced
 - 2UF₆ + Ni → 2UF₅ + NiF₂
 - Other reduction products include U₂F₉, U₄F₁₇, UF₄
- Is frequently chemisorbed
- Is unreactive toward H₂, N₂, O₂ at ambient temperature
- Reacts with many organic materials
 - Attack is most vigorous at the functional group (exceptions: ethers and tertiary amines)
 - Fully fluorinated materials are quite resistant at moderate temperatures
 - Condensed phase reactions can be vigorous
- Is soluble in materials with which it does not react
- Destroys silicones

(Teflon TFE) and copolymers of tetrafluoroethylene and hexafluoropropene (Teflon FEP).

In addition to the rare gases, a few other gases of significance, including N₂, O₂, and H₂, are unreactive with UF₆ at room temperature. Advantage has been taken of this in some separation processes.

Regarding reactivity with organic materials, when attack occurs, it most often starts on the functional groups. Only the tertiary amine, C-N=(C)₂, and the ether, C-O-C, bonds are as resistant to rupture on fluorination as the C-C bond. Organic materials are significantly less reactive with UF₆ vapor than with UF₆ liquid. Hydrocarbons react in a controllable fashion with UF₆ vapor but may react with explosive violence with liquid UF₆.

Fully fluorinated materials such as Teflon TFE, Teflon FEP, and polyhexafluoropropene oxide (Krytox) and similar materials are essentially resistant to fluorination at the temperatures normally employed for handling UF₆ as a liquid. It should be noted that UF₆ has a tendency to dissolve in these materials and others with which it is unreactive or only slowly reactive. In doing so, it obeys the normal principles governing the solubility of one substance in another.^{2,8} One special class of materials, the silicones, are recognized as exhibiting excellent resistance to oxidation in oxygenating atmospheres to relatively high temperatures; and they are not at all stable in UF₆, with which they react to form UO₂F₂ and substituted fluorosilanes.

Toxicity of UF₆ (ref. 9)

Elemental uranium is a highly toxic material on an acute basis. Uranium hexafluoride vapor is toxic *per se*, producing some kidney damage and hydrolyzing to produce HF, which is itself a regulated toxic substance. The threshold limit value (TLV), which is the allowable 8-hour exposure level for industrial workers, is 3 ppm for HF. This value of the TLV for HF translates into a TLV for UF₆ of 0.75 ppm (volume basis) based on the fact that one UF₆ molecule produces four molecules of HF as the essentially instantaneous hydrolysis in the atmosphere occurs. Fortunately, the UO₂⁺⁺ ion has a short biological half-life, and any UO₂F₂ absorbed through the lungs or injected orally is rapidly eliminated from the body by kidney action, thus minimizing the damage.

HANDLING RULES: SOME DO's AND DON'T's WITH UF₆

A few rules for handling UF₆ can be based on the implications of the properties of the compound:

1. Handle UF₆ in a sealed system having vacuum capability to aid in transfers. Liquid transfers are possible in a system that can be operated above about 1.5 atm (0.152 MPa) and 64.05°C (337.20 K).
2. Keep UF₆ away from moisture; otherwise, it will be lost from the gas phase as UO₂F₂.
3. Don't breathe UF₆ or its reaction products. Get respiratory protection before handling heated containers; handle them preferably in a fume hood.
4. Leave at least 40% ullage in cold traps.
5. Don't hook UF₆ cylinders directly to vacuum pumps.
6. Remember that liquid UF₆ and organic materials, other than fluoroplastics, can react violently. (Do not heat cylinders known to contain UF₆ and liquid hydrocarbons.)

SUMMARY

In summary, be reminded that the properties of UF₆ determine how it must be handled and make direct observation impossible. To determine that the material in a container is UF₆, one must use other instruments in addition to a scale. Because of the very large volume expansion of UF₆ upon melting, diligence must be exercised in filling cylinders in which the UF₆ is partially solidified. A cylinder of liquified UF₆ with no ullage is potentially the equivalent of a superheated hot water heater, not just a hydraulically overpressurized cylinder. Finally, UF₆ can be handled safely by careful attention to the suggested precautions.

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ACUTE TOXICITY OF URANIUM HEXAFLUORIDE, URANYL FLUORIDE AND HYDROGEN FLUORIDE

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ABSTRACT

Uranium hexafluoride (UF_6) released into the atmosphere will react rapidly with moisture in the air to form the hydrolysis products uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF). Uranium compounds such as UF_6 and UO_2F_2 exhibit both chemical toxicity and radiological effects, while HF exhibits only chemical toxicity. This paper describes the development of a methodology for assessing the human health consequences of a known acute exposure to a mixture of UF_6 , UO_2F_2 , and HF.

1. INTRODUCTION

Uranium hexafluoride (UF_6) released into the atmosphere will react rapidly with moisture in the air to form the hydrolysis products uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF). The corrosive HF vapor formed by this reaction has a pungent odor and is very irritating to the skin and mucous membranes. The soluble uranium compounds, UO_2F_2 and UF_6 , exhibit both chemical toxicity and radiological effects.

Individuals exposed to these toxic materials may suffer varying health effects depending upon the concentration of the toxicant, the duration of the exposure and many other factors. For example, an accident evaluation may require consideration of the ability of personnel to escape quickly, the variation in the spatial concentration of the toxicant(s), and the physical activity level at the time of exposure. Although these factors may be important when evaluating the hazard associated with an accidental UF_6 release, the information presented in this report does not attempt to account for all the many variables that may need to be considered in a hazard evaluation. Rather, this report focuses on predicting the health effects given the exposure duration and toxicant concentration.

2. CHEMICAL TOXICITY OF UF_6 , HF and UO_2F_2

In 1980 a group of experts in the field of chemical toxicity of soluble uranium and HF were asked to apply known data and make their best

judgments about the toxicological effects of postulated exposures to soluble uranium and HF. This information was then used to develop preliminary Design and Analysis guidelines for estimating the toxicity of soluble uranium and HF.

A review of the information obtained during the development of the preliminary Design and Analysis guidelines indicated a lack of directly applicable data for assessing the consequences of acute UF_6 , UO_2F_2 and HF exposures. Therefore, it was concluded that it would be desirable to obtain additional data on the consequences of acute exposures to UF_6 and UO_2F_2 hydrolysis products. The U. S. Department of Energy (DOE) sponsored a series of animal toxicity experiments at the University of Rochester in order to provide additional data to support accident assessments of U. S. UF_6 handling facilities.

The primary objective of the toxicity experiments was the development of a procedure for evaluating the consequences of acute exposures to mixtures of UF_6 and UO_2F_2 hydrolysis products. This goal was achieved by completing the following tasks:

- (1) determination of the lethal exposure susceptibility of rats and guinea pigs to UF_6 and the UF_6 hydrolysis products UO_2F_2 and HF;
- (2) definition and measurement of delayed effects of uranium and fluorine in animal survivors of UF_6 , UO_2F_2 and HF exposures; and
- (3) prediction from the results of the animal experiments, minimum exposure levels for humans of UF_6 and UO_2F_2 /HF mixtures which will result in significant physiological damage for short periods of exposure.

In late 1983 the experimental work was completed and documented in a report submitted by the University of Rochester to Martin Marietta Energy Systems, Inc.(1) As indicated in Table 1, a total of 66 experiments were conducted utilizing 511 rats and 78 guinea pigs for exposure durations ranging from 2 to 60 minutes. The results of the University of Rochester rat and guinea pig experiments are summarized in Table 2.

A "Delphi" panel of toxicologists was formed to interpret the experimental results. J. B. Hursh, L. J. Leach, and P. E. Morrow of the University of Rochester and M. E. Wrenn of the University of Utah were asked to develop independent, preliminary estimates of the toxicity of UF_6 hydrolysis products. These preliminary toxicity estimates were presented at a meeting where each

¹Operated by Martin Marietta Energy Systems, Inc. for the U. S. DEPARTMENT OF ENERGY under Contract No. DE-AC05-84OR21400.

Table 1. Summary of University of Rochester Animal Experiments

No. and Species	No. of Experiments	Exposure Duration (min)	Air Concentration in g-U/m ³ - Mortality Relationship			
			LC50 ^(a)	95% Confidence Interval	LC10 ^(b)	95% Confidence Interval
150 Rats	15	2	120.0	99.3 - 140	55.0	40.0 - 76.0
170 Rats	17	5	38.6	26.8 - 55.7	10.5	6.48 - 17.1
140 Rats	14	10	12.0	10.1 - 14.3	5.16	3.65 - 7.29
51 Rats	7	60	0.74	0.49 - 1.10	0.27	0.12 - 0.47
78 Guinea Pigs	13	2	62.1	43.4 - 88.8	13.5	5.45 - 33.5

(a) Concentration corresponding to 50% lethality.

(b) Concentration corresponding to 10% lethality.

Table 2. Dose-Response Relationships for Rats and Guinea Pigs

Species	50% Lethal Concentration (g-U/m ³)	Exposure Duration (min)	Concentration-Time Product (g-U/m ³)(min)
Rat	120	2	240*
Rat	38.6	5	193
Rat	12.0	10	120*
Rat	0.74	60	44
Guinea Pig	62.1	2	124

*These numbers are statistically different and indicate that in the rat studies the concentration-time product does not have a constant value for a given biologic response (percent mortality).

toxicologist presented his approach and rationale for estimating the toxicity associated with acute UF₆, UO₂F₂ and HF exposures. The toxicologists were then asked to reevaluate their toxicity estimates, if necessary as a result of the discussions, and to submit documentation describing the rationale used in developing their "final" estimates of toxicity. The final toxicity estimates were then used to develop a basis for assessing the toxicity of soluble uranium and HF.(2)

The basis derived by the panel of toxicologists for assessing the toxicity of soluble uranium compounds such as UF₆ and UO₂F₂ is presented in Table 3 and on Figure 1. Four health effect classifications were established for characterizing the toxicity of soluble uranium compounds: (1) no effect, (2) possible mild health

effects, (3) renal injury, and (4) lethality.

The possible mild health effects regime corresponds to exposure levels that are greater than the no effect level and less than the renal injury level. An exposure predicted to result in possible mild health effects may result in observable biological effects, but such exposures will not, in themselves, result in either a short term or a long term impairment in the body's ability to function. The renal injury classification indicates that significant physiological damage to the kidneys is predicted. The lethality health effect classification corresponds to an exposure that is expected to result in 50% mortality. Estimates of uranium toxicity for exposure times greater than 60 minutes should be based on extrapolation of the 60-min toxicity estimates as shown on Figure 1.

Table 3. Chemical Toxicity of Soluble Uranium Compounds

Health Effect	Uranium Absorption ^a (mg-U/kg)	Exposure Level ^b (mg-U/m ³)(min)	Exposure Time (min)
50% Lethality	1.63	35,000	Indefinite
Renal Injury	0.058	1,250 750	≤ 30 min ≥ 60 min
No Effect	0.03	650 390	≤ 30 min ≥ 60 min

^aThe absorbed quantity of uranium per kg of body weight.

^bThe exposure level is defined as the product of the airborne and the exposure time. Based on the ICRP resting respiration rate of 7.5 L/min.

Estimates of HF exposure levels as related to various physiological effects are presented in Table 4 and on Figure 2. Five health effect classifications have been established for exposures to HF: (1) no effect, (2) smell/no health effects, (3) smell/possible irritation, (4) irritation/possible health effects, and (5) lethal. An HF exposure is predicted to have no effect if the HF concentration is less than the detection by smell level.

3. RADIOTOXICITY OF URANIUM

Uranium-234, uranium-235, and uranium-238 are alpha emitters of widely varying specific activity. U-234 has a specific activity approximately 2900 times that of U-235 and 18,000 times that of U-238. Thus, U-234 has the greatest radiotoxic potential. For example, at 3% U-235 enrichment, the U-234 represents only 0.222 wt.% of the total uranium present, but it is

responsible for more than 78% of the total activity and, hence, total radiation dose.

Based on experiments in which small animals were subjected to plutonium exposure, it appears that the minimum health effect resulting from acute exposure to alpha radiation is a decreased immune response, which could occur when the total activity in the lung exceeds 100 μCi .⁽³⁾

Table 5 depicts the relative chemical toxicity and radiotoxicity of uranium that has been enriched to 97.5% U-235 and 1.14% U-234. It is evident, from the data shown in Table 5, that for acute exposures to soluble uranium, radiotoxicity is negligible in comparison with chemical toxicity, even at 97.5% U-235 enrichment and 1.14% U-234 enrichment. Therefore, it is concluded that the radiotoxicity of an acute uranium exposure is insignificant when compared with the associated chemical toxicity.

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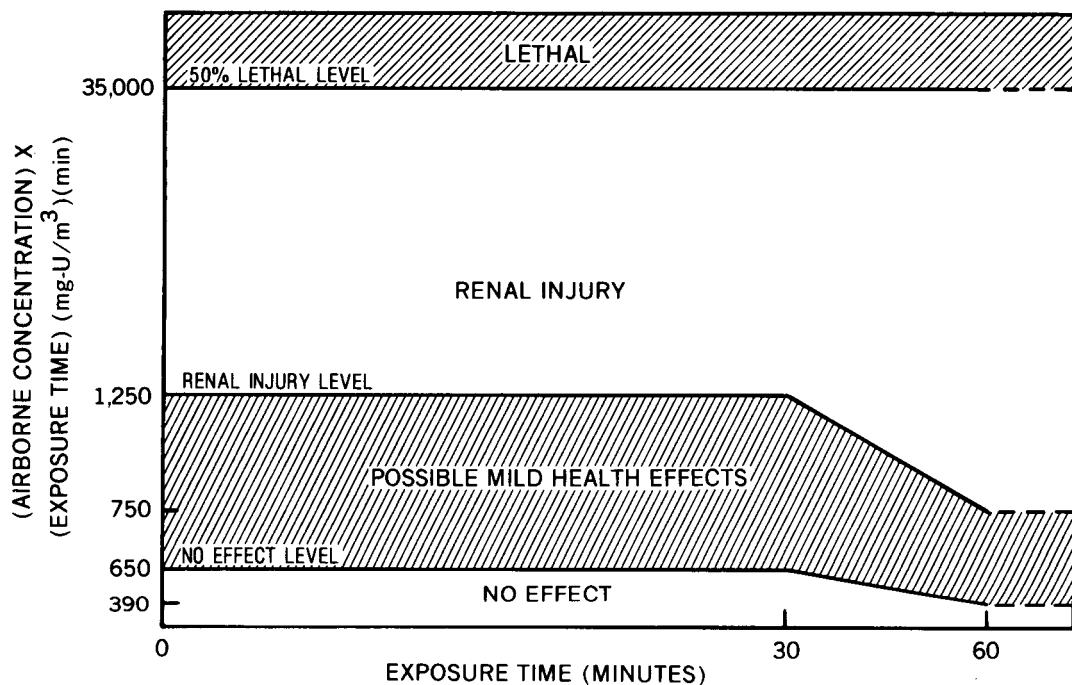


Figure 1. Toxicity of Acute Exposures to Soluble Uranium.

Table 4. Toxicity of Hydrogen Fluoride

Health Effect	HF Concentration (mg/m ³)	Exposure Time
Detection by Smell	1	Indefinite
Smell/No Health Effects	2.5	≤ 8 hr
Irritation	26	< 10 min
	13.33	≥ 10 min
Lethal*	53,000	0 to 60 min
	exposure time (min)	

*Estimates of HF lethality are based on an inhaled exposure of 53,000 (mg-HF/m³)(min).

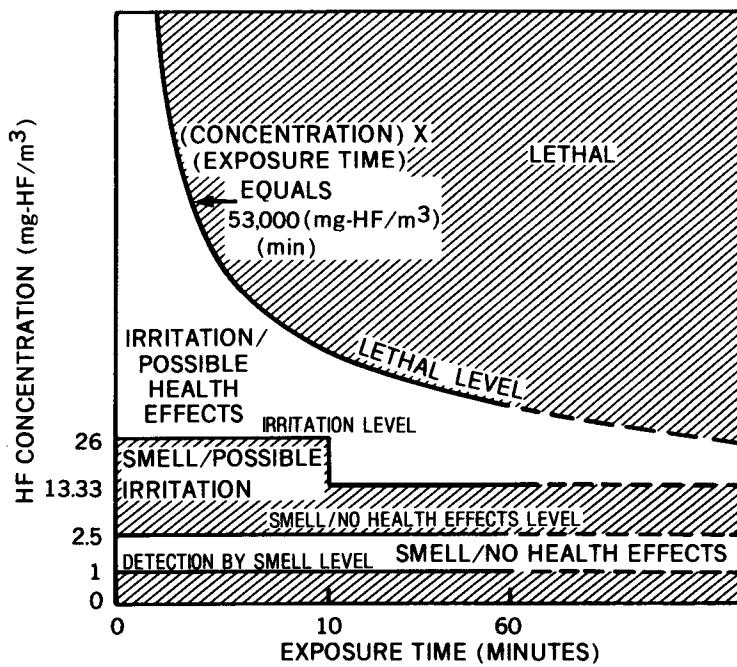


Figure 2. Toxicity of Acute Exposures to Hydrogen Fluoride.

Table 5. Comparison of Chemical Toxicity and Radiotoxicity of Soluble Uranium*

Absorbed Dose of Soluble Uranium (mg-U/kg)	Equivalent Radiation Dose (μCi)	Acute Health Effects	Chemical Toxicity	Radiotoxicity
0.03	0.16	No effect	No effect	
0.058	0.30	Renal injury	No effect	
1.63	8.45	50% lethality	No effect	
19.29	100.	Lethal	Onset of radiological effects	

*At 97.5% U-235 and 1.14% U-234 enrichment.(4)

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An Experimental Study on Heat Transfer of A UF_6 -Filled Vessel

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ABSTRACT

Thermal tests for a bare vessel filled with uranium hexafluoride were conducted at Tokai Works of PNC during August 1987, for a better understanding of heat transfer phenomena in a hex cylinder. The equipment for thermal tests consists of a 270 mm in diameter, 1400 mm long and 30 mm thick carbon steel test cylinder encased with a 20 kW electric heater and measuring sensors. The test cylinder was filled with about 110 kg of uranium hexafluoride, which amounted to 95% in volume of the test cylinder when the temperature of inner uranium hexafluoride became 120°C. The temperature of the heater surface was controlled by a PID controller in the range from 80 to 400°C. The setup was equipped with a capacitance manometer to measure inner pressure of the test cylinder and 28 conventional seathed thermocouples attached to various places of both the equipment and inner uranium hexafluoride, which allowed us to observe the phase changes of inner uranium hexafluoride as a function of time. Based on the experimental observation, we proposed a heat transfer model suitable for numerical analysis and derived the values related to heat transfer in a cylinder filled with uranium hexafluoride by using a TRUMP code.

INTRODUCTION

The main purpose of thermal tests for a bare vessel filled with uranium hexafluoride is to derive values related to the heat transfer, such as the heat transmitted to inner uranium hexafluoride, the heat transfer coefficient between the cylinder material and uranium hexafluoride, and the apparent thermal conductivities of solid and liquid uranium hexafluoride. Another purpose is to observe the phase changes of uranium hexafluoride in the test cylinder as a function of time to construct a heat transfer model suitable for numerical analysis. These experimental findings will contribute to determine analytically if the 48Y cylinders would hydrostatically rupture and the time available for fire fighting before the incident occurred.

EXPERIMENTAL

Shown in Fig.1 is an equipment for the thermal tests, which consists of a 270 mm in diameter, 1400

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mm long and 30 mm thick carbon steel test cylinder encased with a 20 kW electric heater and measuring sensors. The test cylinder has a valve on its end plate to imitate an actual cylinder and was filled with about 110 kg of uranium hexafluoride, which amounted to 95% in volume of the test cylinder when the temperature of inner uranium hexafluoride became 120°C. The heater was encased by the reflector and the insulator to maintain uniform heating. This heating apparatus was controlled by a PID controller in the temperature range from 80 to 400°C on the heater surface.

As for the measuring sensors, there were 28 conventional seathed thermocouples to measure temperatures of various places of the equipment and a MKS 5034 capacitance manometer to measure inner pressure of the test cylinder. Among 28 thermocouples, two sets of five thermocouples were for temperature measurements of the heater surfaces and the valve located on the end plate of the cylinder. Temperatures of the outer and inner surfaces as well as temperatures of uranium hexafluoride

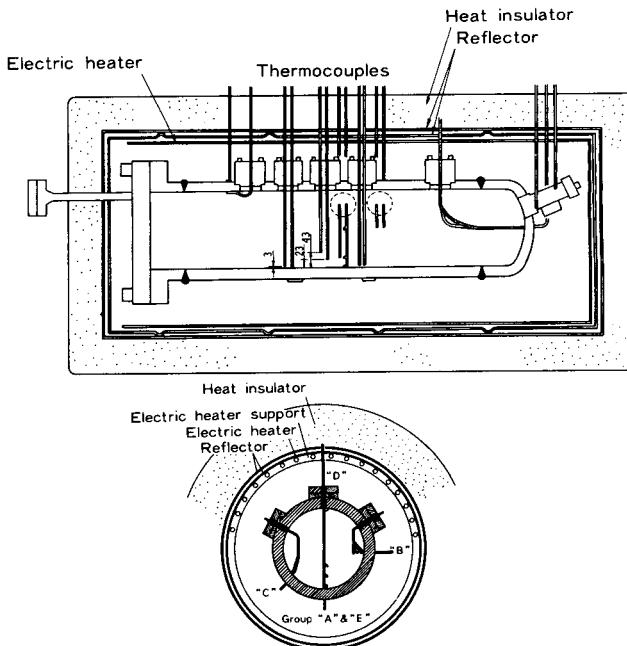


Fig.1 The test cylinder encased with electric heater, reflector and heat insulator. The figure also shows thermocouples for temperature measurement which are divided into several groups indicated by "A", "B", "C", "D" and "E".

in the cylinder were measured by the rest 18 thermocouples. These 18 thermocouples were divided into several groups to measure the radial temperature distribution. We labeled the groups measuring the lower and the upper parts on the vertical center line as "A" ("E") and "D", respectively. Labels "B" and "C" were used for the groups measuring the parts with angles of 90° and 45° to the vertical center line. These labels had additional suffices to indicate radial positions. The outer and inner surfaces of the test cylinder were indicated by the suffices "1" and "2" respectively. Indicated by "3", "4", "5" were 3 mm, 23mm and 43mm inwards away from the inner surface.

Fig.2 shows the safety system of the test equipment, which consists of a large volume pressure release tank along with a safety valve and a rupture disc whose working pressures are 3.2 and 4.2 kg/cm², respectively. This figure also shows the data logger using a PC 9801 personal computer which facilitates data acquisition of all the 29 measurements in every three seconds.

Because of its large capacity, the heater temperature went up rather fast and reached the desired level in 7 to 8 minutes after supplying power within the temperature range from 200 to 400°C. In the mean time, we observed that the pressure reached and stayed at the triple point of uranium hexafluoride in a certain period and then rose rapidly. Since we observed that the inner pressure reached and stayed at the triple point vapor pressure of uranium hexafluoride in a certain period and then rose rapidly, we determined the timing to turn off the heater switch after repeated trial operations of the equipment.

During August 1987, the total of 11 runs of thermal tests were carried out as shown in Table 1. In Case 1, the initial state of solid uranium hexafluoride was considered to be in a cylindrical form, because it was the state right after 110 kg of gaseous uranium hexafluoride was first transferred into the cold test cylinder. Tests made after liquefying and purifying uranium hexafluoride are the Cases 2, 3, and 4, where we set the maximum temperature of the heater for 200, 300 and 400°C, respectively.

RESULTS

Shown in the above of Fig.3 is a typical example of the results in our thermal tests. We can see that even after the heater surface reaches the predetermined temperature, 200°C in this case, the inner pressure goes up rather slowly and then shows

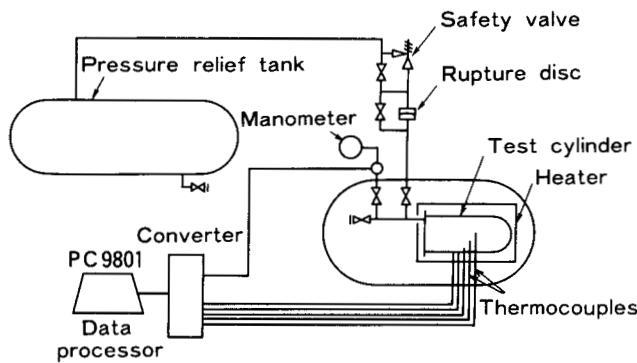


Fig.2 Safety devices for thermal tests and data acquisition system using a PC9801 processor which allows to acquire 29 measurements in every 3 seconds.

Table 1. Conditions of thermal tests carried out in August 1987.

Case No.	UF ₆ condition weight (kg)	initial form	Thermal condition		
			heater temp. (°C)	heating rate (°C/min)	heating time (min.)
1	112	cylinder	400	36	43
2 - 1					42
2 - 2	107	cylinder	200	27	56
2 - 3					60
2 - 4					60
3 - 1		cylinder			26
3 - 2	107	cylinder	300	36	21
3 - 3					20
4 - 1		cylinder			10
4 - 2	107	cylinder	400	36	17
4 - 3					25

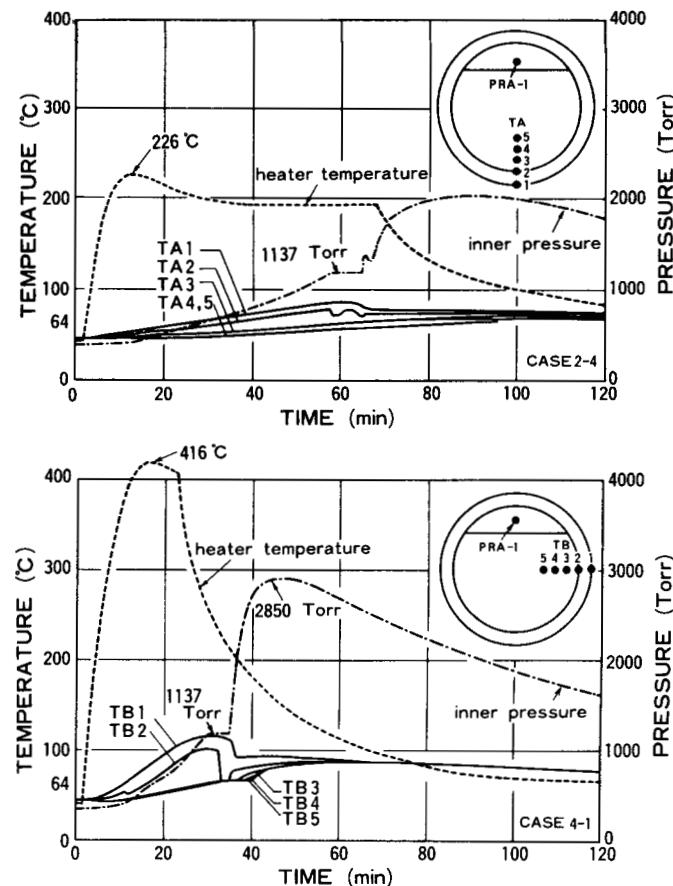


Fig.3 Typical examples of experimental results. The above figure shows both the inner pressure change and the temperature changes as a function of time in Case 2-4. The result in Case 4-1 is shown below. Temperature measurements were shown at different positions from each other.

a plateau at 1137 Torr, which is in good agreement with the triple point vapor pressure of uranium hexafluoride and thus seemed to be the indication of the onset of liquefying. After staying here in some period, the pressure rises rapidly far beyond the triple point, while uranium hexafluoride in a lower part of the test cylinder is considered to remain still in solid state judging from the temperature of TE-3, which is lower than the triple point temperature of uranium hexafluoride, 64°C. The solid uranium hexafluoride in this region gradually melts a while later. One more to be mentioned is the temperature drop of the inner surface right after the inner pressure reaches the triple point vapor pressure of uranium hexafluoride. This trend is much clearer in the bottom part of the test cylinder, whose temperature shows a sharp decrease to 64°C, as shown in TA and TE. This temperature drop is considered to be caused by liquid uranium hexafluoride flowing down in the bottom.

The result for a 400°C operation is shown below in the same figure. Only 10 minutes' holding time caused a subsequent inner pressure increase up to 2850 Torr, which was very close to the rupture disc limit. We can see that the TB's located near the upper surface of uranium hexafluoride read the melting point temperature of uranium hexafluoride when the inner pressure reaches the triple point vapor pressure. Furthermore, only the temperature of TB3 starts to increase immediately after the pressure rises beyond the triple point vapor pressure, while temperature increase of TB4 and TB5 are observed a while later.

In Fig.4, measured temperature changes are plotted along with temperature values calculated from the inner pressure based on the vapor pressure equation of uranium hexafluoride(1). Fairly good agreement is only below the triple point temperature between TB3 and the one calculated from the inner pressure. It should be mentioned that the temperatures of uranium hexafluoride at TB2 and TB3 remain to be far lower than the equilibrium temperature corresponding to the inner pressure. This may mean that the inner pressure reflects only the temperature of a thin layer of liquid uranium hexafluoride surface and thus there is a temperature gradient in uranium hexafluoride which suppresses the natural convection effect.

Fig.5 shows measured temperature distribution

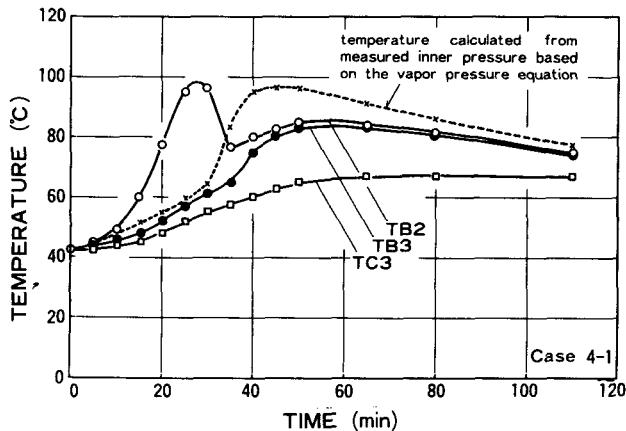


Fig.4 Comparison of temperatures measured near the inner surface of the test cylinder with the temperature calculated from inner pressure based on the vapor pressure equation.

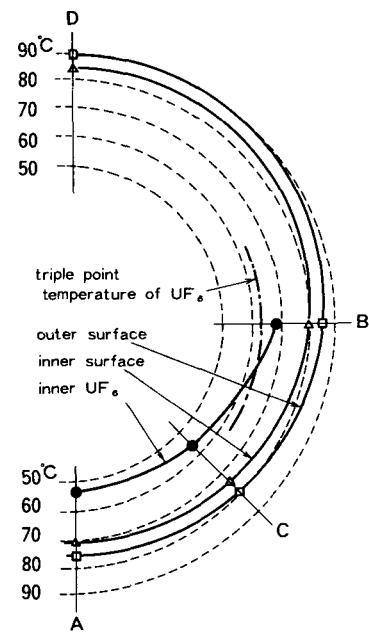


Fig.5 Measured temperature distribution at $t=60$ minutes in Case 2-1. The circle indicates the temperature of uranium hexafluoride measured at the position 3mm inwards away from the inner surface of the test cylinder. The square and the triangle are the temperature of outer and inner surface, respectively.

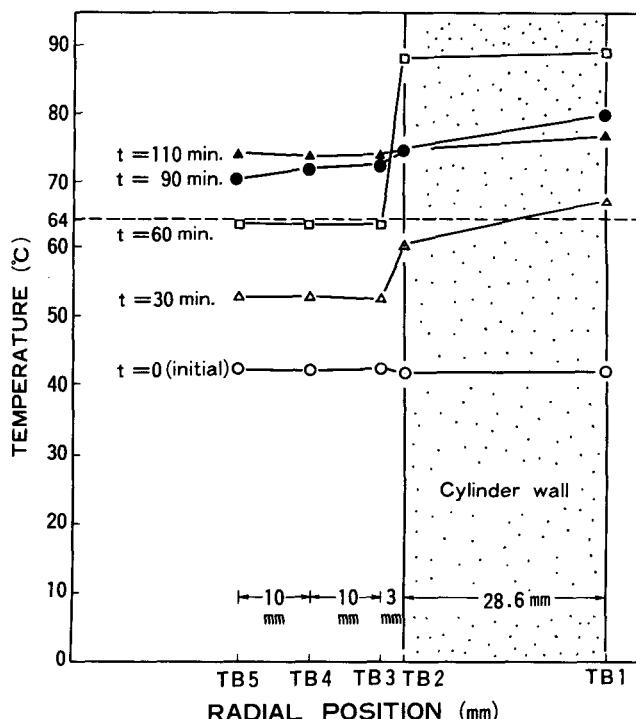


Fig.6 Radial temperature distribution measured by the group B thermocouples. The figure shows its change with the course of time.

at $t=60$ minutes in Case 2-1. The circles, triangles and squares indicate the temperatures of the outer and inner surfaces and 3mm inwards position from the inner surface, respectively. The temperature scale is expressed in terms of the radial length. TC's are plotted on the assumption of axial symmetry. It should be noted that the temperature of liquid uranium hexafluoride can increase over the triple point even when there exists solid phase in the cylinder.

Fig.6 shows the radial temperature distribution changes measured at TB's in the experiment of Case 2-4. When the inner uranium hexafluoride temperature remains lower than its triple point temperature, the temperature difference between the cylinder material and inner uranium hexafluoride increases with time. This leads us to assume a gap conductance between the inner surface of cylinder and uranium hexafluoride. This figure also shows that the temperature difference disappears between the cylinder material and uranium hexafluoride when uranium hexafluoride turns into liquid. This suggests liquid uranium hexafluoride eliminates this gap conductance to absorb the enthalpy of cylinder material.

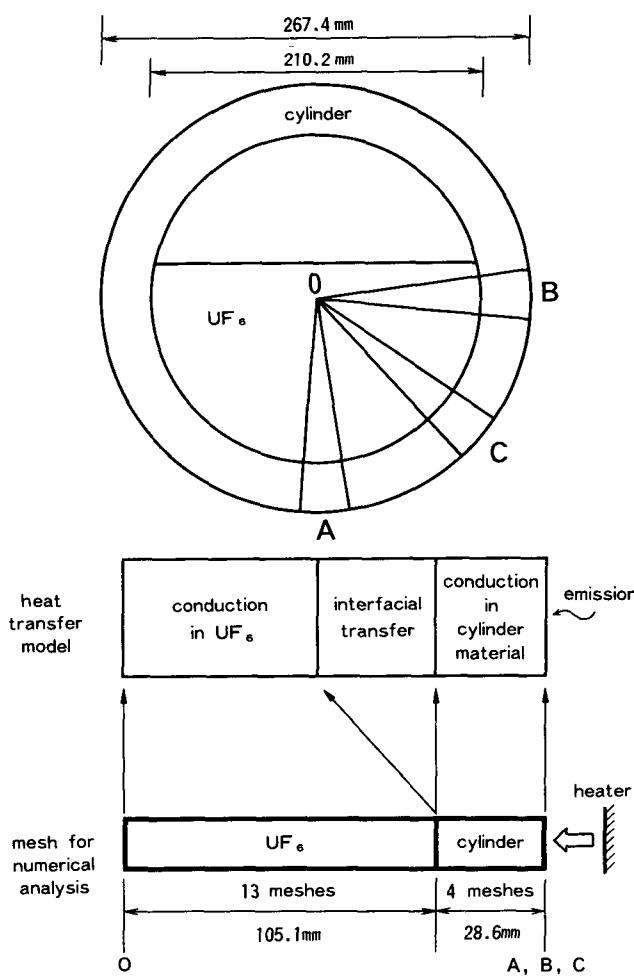


Fig.7 Heat transfer model and mesh division used in one dimensional analysis.

Table 2. Overall emissivity on the cylinder surface and heat transfer coefficient between inner surface of the cylinder and uranium hexafluoride, derived through one dimensional analysis using the experimental data.

Direction (θ)	heat transfer coefficient ($\text{kcal}/\text{m}^2 \text{h}^\circ\text{C}$)								overall emissivity (ϵ)	
	temperature ($^\circ\text{C}$)									
	0	53	60	62	64	70	72	85		
A	11	11	19	27	74	—	—	—	0.55	
B	15	15	23	25	30	210	330	1150	0.60	
C	10	10	17	22	52	—	—	—	0.58	

Table 3. Apparent heat conductivity of uranium hexafluoride obtained by one dimensional analysis.

Description	apparent heat conductivity of UF_6 ($\text{k cal}/\text{m h}^\circ\text{C}$)							
	temperature ($^\circ\text{C}$)							
	35	45	55	59	63	64	65	
This work	0.05	0.73	0.97	1.23	1.80	2.25	—	250
PATRAM '83	—	—	2.58	—	—	—	3.44	—

DISCUSSION

The hydrostatic rupture concept of the cylinder caused by the expansion of liquid uranium hexafluoride under fire was usually based on the following. The heat entering through the cylinder wall would make all the uranium hexafluoride melt away in an equilibrium state at the triple point. And it would be not until the whole uranium hexafluoride became liquid that the inner pressure and the temperature of liquid uranium hexafluoride started to increase over the triple point ones. However, the results of our experiment indicates the different features as stated in the preceding paragraph.

Prior to making a two-dimensional analysis, the estimation of values related to heat transfer was carried out by an one-dimensional analysis. In this one-dimensional analysis, as shown in Fig.7, the emission of heat between the surfaces of heater and cylinder was expressed in terms of an overall emissivity and we assume an interfacial heat transfer model to take account of the gap conductance due to a gaseous layer resulting from the sublimation of uranium hexafluoride near the inner surface of the test cylinder by using a heat transfer coefficient. 4 and 13 meshes were used for the calculation of heat conduction in the cylinder material and in uranium hexafluoride, respectively. The calculation for an overall emissivity and a heat transfer coefficient was carried out for three regions of the test cylinder as indicated by A, B and C in Fig.7. The results obtained by iterative simulations with the use of a TRUMP code are given in Tables 2 and 3. The overall emissivity obtained here is very close to 0.6 which is a little smaller than the value recommended by IAEA. The heat transfer coefficient has reasonable reflection of the gap conductance change observed in the experiment. As for the apparent heat conductivity of uranium hexafluoride,

a little lower values are obtained in comparison with those reported by Duret and Bonnard(2). The origin of this difference will be clarified by another test program under way in our facility for the measurement of heat conductivities of solid and liquid uranium hexafluoride by both equilibrium and non-equilibrium methods.

Fig.8 shows a heat transfer model and a mesh configuration used in a two-dimensional analysis. This configuration represents only a right half of the test cylinder cross section based on a symmetric assumption. The mesh division is intended to be suitable for heat flux calculation. Another to be mentioned is finer meshes near the inner surface

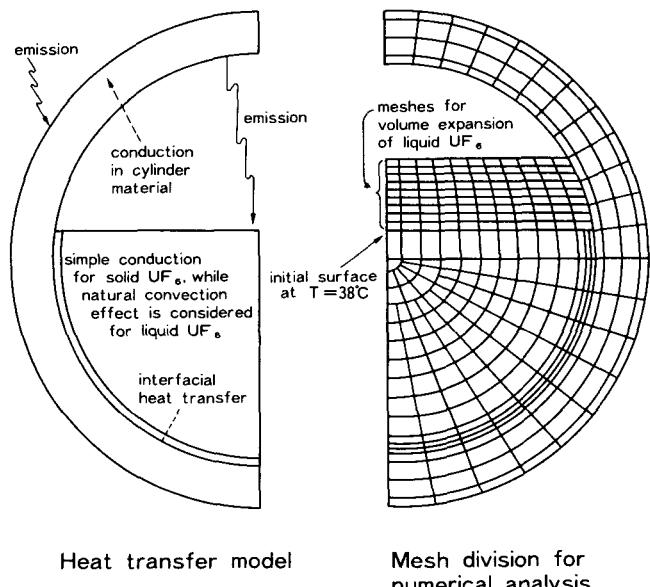


Fig.8 Heat transfer model and mesh division used in 2-dimensional analysis.

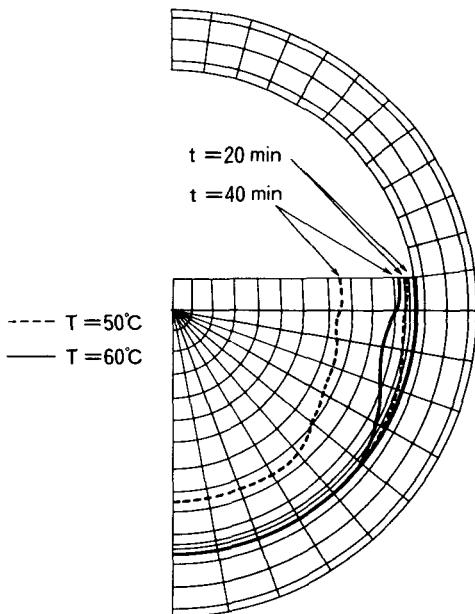


Fig.9 Temperature of inner uranium hexafluoride calculated for the thermal test of $T=400^{\circ}\text{C}$ on the heater surface.

to take account of the natural convection as well as prearranged meshes to accomodate the volume expansion of liquid uranium hexafluoride. The calculation was made by using a TRUMP code slightly modified by us.

Shown in Fig.9 is the simulation result of inner uranium hexafluoride temperature corresponding to the thermal test of $T=400^{\circ}\text{C}$ on the heater surface. The result has a good reproducibility of the experimental trend as shown in Fig.5. The calculated temperature change of uranium hexafluoride is shown in Fig.10. For comparison, experimental temperatures are also plotted in the same figure. In spite that the calculation is made with the use of the heat transfer values obtained through an one-dimensional analysis, a fairly good agreement is obtained between the calculation and the experiment. The reason why the agreement becomes poor at higher temperature is due to the negligence of the latent heat of vaporization of uranium hexafluoride. Modification must be made in computing the heat transfer problem of a 48Y cylinder at 800°C .

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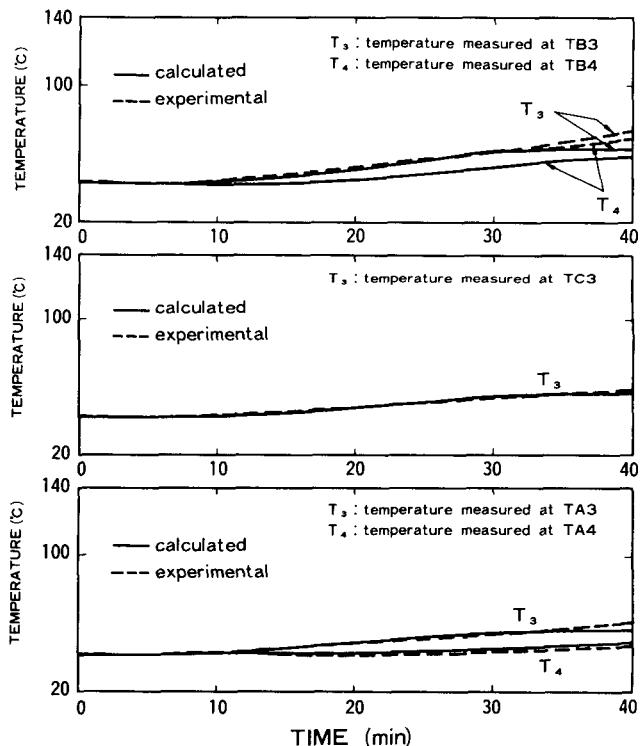


Fig.10 Time dependence of inner uranium hexafluoride temperature simulated for the thermal test of $T=400^{\circ}\text{C}$ on the heater surface by 2-dimensional analysis with the use of heat transfer values listed in Tables 2 and 3.

INVESTIGATION OF UF₆ BEHAVIOR IN A FIRE¹

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ABSTRACT

Reactions between UF₆ and combustible gases and the potential for UF₆-filled cylinders to rupture when exposed to fire are addressed. Although the absence of kinetic data prevents specific identification and quantification of the chemical species formed, potential reaction products resulting from the release of UF₆ into a fire include UF₄, UO₂F₂, HF, C, CF₄, COF₂, and short chain, fluorinated or partially fluorinated hydrocarbons. Such a release adds energy to a fire relative to normal combustion reactions. Time intervals to an assumed point of rupture for UF₆-filled cylinders exposed to fire are estimated conservatively. Several related studies are also summarized, including a test series in which small UF₆-filled cylinders were immersed in fire resulting in valve failures and explosive ruptures. It is concluded that all sizes of UF₆ cylinders currently in use may rupture within 30 min when totally immersed in a fire. For cylinders adjacent to fires, rupture of the larger cylinders appears much less likely.

NOMENCLATURE

A	=	area, ft ²
E	=	total heat requirements for heating a cylinder and UF ₆ from initial to final conditions, Btu
F	=	view factor
AH	=	enthalpy change from initial to final conditions, Btu/lb
P	=	pressure, psia

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²Operated by Martin Marietta Energy Systems, Inc., for the U.S. Department of Energy under Contract No. DE-AC05-84OR21400.

q ₂	=	heat flux relative to the cylinder surface area, Btu/h·ft ²
Q	=	heat rate, Btu/h
r	=	cylinder radius, in
t	=	wall thickness, in
T	=	absolute temperature, °R
ε	=	emissivity
σ	=	Stefan-Boltzman constant
	=	0.173 x 10 ⁻⁸ Btu/h·ft ² ·°R ⁴
σ _u	=	ultimate stress, psia
τ	=	time to rupture, min
1,2	=	subscripts denoting fire and cylinder, respectively

INTRODUCTION AND SUMMARY

In 1985, the Nuclear Regulatory Commission (NRC) requested that consideration be given to several UF₆-fire issues as a part of an ongoing program to develop an Accident Analysis Handbook. The issues concern (I) the reactions occurring between UF₆ released into a fire and combustible gases and combustion products and (II) the potential for UF₆-filled cylinders to rupture when exposed to fire. The results presented in this paper represent the current status of investigation into these issues.

Potential reaction products resulting from the release of UF₆ into a fire include UF₄, UO₂F₂, HF, C, CF₄, COF₂, and short chain, fluorinated or partially fluorinated hydrocarbons. UF₆ reactions with combustible gases add energy to a fire relative to normal combustion reactions with O₂. However, energy release appears to be maximized by the complete combustion of hydrocarbons to H₂O and CO₂ along with the complete hydrolysis of UF₆ by H₂O. The absence of kinetic data precludes identification of the most likely chemical species resulting from the release of UF₆ into a fire or, consequently, the corresponding energy increase. The development of appropriate kinetic data would require a substantial experimental program.

Time intervals to an assumed point of rupture for UF₆-filled cylinders (liquid UF₆ at 300°F) exposed to fire have been estimated in what should be considered conservative, preliminary calculations. Consideration was given to cylinders fully immersed in a fire and to those adjacent to a fire. Fire conditions utilized in the analyses encompass NRC criteria and a proposed ASTM standard. Several related studies are summarized, including a series of tests in which small UF₆-filled cylinders (corresponding to 5A- and 8A-sized cylinders) were immersed in fire resulting in valve failures and explosive ruptures. It appears reasonable to conclude that all sizes of UF₆ cylinders currently in use may rupture within 30 min when totally immersed in a fire; in some cases, there may be

insufficient time to begin fighting a fire before rupture occurs. For cylinders adjacent to fires, rupture of the larger cylinders (i.e., 30B, 48X, 48Y) appears much less likely.

I. UF_6 -FIRE PRODUCT REACTIONS

The reaction of UF_6 with H_2O , which occurs rapidly in the ambient environment, would also occur in a fire due to the large quantities of H_2O formed from the combustion of hydrocarbons. Free-radical reactions between UF_6 and combustion products would also be favored by the high temperatures of a fire. Possible reaction products include UF_4 , HF , C , CF_x , and COF_2 ; fluorine will also substitute freely into hydrocarbon chains ($-\text{C}_n\text{H}_{2n-}$).⁽¹⁾ Under non-fire conditions, UF_6 and hydrocarbon oils have reacted explosively. Rapp⁽²⁾ described consequent reaction products as "black carbonaceous smoke," "carbon and reduced uranium in the residue," "uranium in the reduced state and an elevated carbon content," "solid residues ... consisted of β UF_5 containing about 4% U_2F_9 in association with a small amount of fluorinated carbonaceous material," and "reduced uranium fluoride." Experimental results indicate that the "reaction between uranium hexafluoride and hydrocarbon oil becomes vigorous at 70 to 90°C, forming UF_4 , carbon, and low molecular weight fluorinated compounds (CF_4 , C_2F_6 , C_3F_8 , C_4F_{10})."⁽²⁾ He further states that "where excess UF_6 is involved the reduced uranium most probably would consist of some UF_5 , U_2F_9 and/or U_4F_{17} ." In the absence of kinetic data, the final chemical species resulting from a release of UF_6 into a fire and the corresponding energy increase cannot be determined. While a few well chosen experiments may provide

useful information, obtaining sufficient data to predict with reasonable accuracy what occurs when UF_6 is released into a fire would require a major experimental program.⁽³⁾

Nevertheless, potential effects of the release of UF_6 into a fire can be evaluated. Several possible reactions involving UF_6 and CH_4 , H_2 , C , and CO --combustible materials chosen as surrogates for the broad range of gases present within a fire--are listed in Table 1 along with combustion reactions (leading to formation of H_2O and CO_2) and the UF_6 hydrolysis reaction. Consideration has been given to energy trade-offs occurring when the surrogate materials (e.g., CH_4 , H_2 , C , CO) react with UF_6 rather than O_2 . Results of this comparison are given in Table 2; in all cases, more heat is released by reacting the surrogates with UF_6 rather than with O_2 . On the other hand, the heat of reaction for UF_6 and H_2O is -101.5 kJ/mol UF_6 , which exceeds the increased energy releases tabulated in Table 2. Consequently, energy release into the fire appears to be maximized by complete combustion of hydrocarbons along with the complete hydrolysis of UF_6 .

If a carbon-to-hydrogen ratio approaching 2 (i.e., $-\text{C}_n\text{H}_{2n-}$) is assumed for a fuel contributing to a fire, a simple mass balance yields an off-gas composition of about 13% H_2O assuming dry air for combustion. This composition significantly exceeds ambient concentrations. When UF_6 is released into a fire environment--whether as a sudden, explosive release or in a slower release through a crack, the subsequent flashing and turbulence should yield rapid mixing and reaction of the UF_6 , with either H_2O or combustible materials.

Table 1. Some Possible Reactions between UF_6 and Fire Products^a

Reactions	ΔH_{rxn} , kJ/mol	ΔG_{rxn} , kJ/mol
1. $\text{UF}_6(\text{v}) + 2 \text{ H}_2\text{O}(\text{v}) \rightarrow \text{UO}_2\text{F}_2(\text{s}) + 4 \text{ HF}(\text{v})$	-101.5	-123.8
2. $\text{UF}_6(\text{v}) + 0.25 \text{ CH}_4(\text{v}) \rightarrow \text{UF}_4(\text{s}) + 0.25 \text{ CF}_4(\text{v}) + \text{HF}(\text{v})$	-250.4	-239.9
3. $\text{UF}_6(\text{v}) + \text{H}_2(\text{v}) \rightarrow \text{UF}_4(\text{s}) + 2 \text{ HF}(\text{v})$	-309.4	-306.0
4. $\text{UF}_6(\text{v}) + 0.5 \text{ C}(\text{s}) \rightarrow \text{UF}_4(\text{s}) + 0.5 \text{ CF}_4(\text{v})$	-229.3	-199.1
5. $\text{UF}_6(\text{v}) + \text{CO}(\text{v}) \rightarrow \text{UF}_4(\text{s}) + \text{COF}_2(\text{v})$	-291.0	-241.6
6. $\text{CH}_4(\text{v}) + 2 \text{ O}_2 \rightarrow \text{CO}_2(\text{v}) + 2 \text{ H}_2\text{O}(\text{v})$	-802.3	-800.8
7. $\text{H}_2(\text{v}) + 0.5 \text{ O}_2(\text{v}) \rightarrow \text{H}_2\text{O}(\text{v})$	-241.8	-228.6
8. $\text{C}(\text{s}) + \text{O}_2(\text{v}) \rightarrow \text{CO}_2(\text{v})$	-393.5	-394.4
9. $\text{C}(\text{s}) + 0.5 \text{ O}_2(\text{v}) \rightarrow \text{CO}(\text{v})$	-110.5	-137.2

^a The values of ΔH_{rxn} and ΔG_{rxn} are based on data taken from Ref. 4. Reference conditions are 25°C and 0.1 MPa.

Table 2. Energy Trade-offs for Reaction with UF_6 vs O_2

Reactant	Change in energy released (kJ/mol UF_6)			Net increase in energy release to fire (%)
	$(\Delta H_{\text{rxn}} \text{ w/UF}_6)$	- $(\Delta H_{\text{rxn}} \text{ w/O}_2)$	= Net Change	
CH_4	-250.4	-802.3 / 4	-49.8	25
H_2	-309.0	-241.8	-67.2	28
C	-229.3	-393.5 / 2	-32.6	17
CO	-291.0	-393.5 - (-110.5)	-8.0	3

II. CYLINDER RUPTURE DUE TO FIRE

The time required to rupture a cylinder exposed to fire has been conservatively estimated. Results are compared to experiments conducted in 1965.

FIRE CONDITIONS

There are several sources of fire conditions which may be used for analysis of fire effects. NRC criteria are as follows:(5)

Exposure of the whole specimen for not less than 30 minutes to a heat flux not less than that of a radiation environment of 800°C (1475°F) with an emissivity coefficient of at least 0.9. For purposes of calculation, the surface absorptivity must be either that value which the package may be expected to possess if exposed to a fire or 0.8, whichever is greater. ...

Buck and Belason included the following description of a design fire environment relative to a proposed ASTM standard:(6)

A total heat flux of 174 kW/m² (15.28 Btu/ft²·s) with components of 158 kW/m² (13.89 Btu/ft²·s) radiative heat flux and 16 kW/m² (1.39 Btu/ft²·s) convective heat flux, average flame temperatures of between 983°C (1700°F) and 1261°C (2300°F) ...

They also argue that "in ... large hydrocarbon pool fires, it [is] reasonable to assume an emissivity of 1.0" since "the flames only have to be 3 to 6 feet thick to be optically opaque."

The tabulated results presented subsequently assume a flame temperature of 1475°F and a flame emissivity of 1.0. It is also assumed (for the case of complete immersion in a fire) that the convective heat flux to the cylinder, which would be about 10% of the total heat flux based on the proposed ASTM standard, is negligible relative to other uncertainties.

CYLINDER RUPTURE CONDITIONS

Based on nominal cylinder characteristics (see Table 3), a cylinder containing the maximum quantity of UF₆ would be completely filled with liquid at 300°F. This condition was initially considered as a criterium for imminent rupture;

however, more realistic failure conditions can be extrapolated from data obtained by hydraulically rupturing UF₆ cylinders. Such data are summarized in Table 4.

For cylinders--30B and smaller--that exhibit ductile failure (hoop stress), hydrostatic failure conditions obtained at room temperatures were extrapolated to fire conditions by multiplying the hydrostatic failure pressure and volume increase by a materials degradation factor of 0.35 based on an assumed temperature of 1200°F.(10) This factor was used for both steel and monel; however, a factor greater than 0.35 is more probable for monel (i.e., monel experiences less degradation than steel). Because data were not available for 5A and 8A cylinders, the following relation for determining failure pressure was used:

$$\sigma_u = P r/t . \quad (1)$$

In this instance, σ_u was calculated from the failure pressure of a 12B cylinder, then failure pressures were evaluated for the smaller cylinders. The volume increase of 5A and 8A cylinders was assumed to be the same as that of a 12B cylinder.(11)

The failure mechanism for 10- and 14-ton cylinders is brittle fracture: the stiffening rings develop cracks where the ends are welded together that propagate inward through the tack weld joining the rings to the cylinders. If the stiffening rings were not present, the volume increase of these cylinders is expected to be comparable to that of the 30B cylinders. For these 10- and 14-ton cylinders, failure pressure at fire conditions was determined from Eq. 1 based on failure conditions for 30B cylinders; however, the volume increase was only slightly reduced from that determined from the hydrostatic rupture tests. The rationale for this approach is that brittle failure is not accelerated by higher temperatures, but there is a potential for a greater volume increase, up to about 10%, from hoop stress prior to failure. Assuming only a slight reduction in volume increase is therefore considered reasonable.(12)

Given estimates of the failure pressure and final volume of UF₆ cylinders, the final temperature of UF₆ can be estimated from physical property correlations for liquid density, compressibility, and vapor pressure. Estimated conditions for UF₆ cylinder failure in a fire are also presented in Table 4. The total heating requirements, from a range of initial conditions (solid UF₆ at 70°F through liquid UF₆ at 225°F), to the final rupture

Table 3. Cylinder Characteristics^a

Type	Tare weight, lb	Maximum capacity, lb	Internal volume, ft ³	Internal diameter, in	Average length, in	Surface area, ft ²
5A	55	55	0.284	5	24.99	3.00
8A	120	255	1.319	8	45.34	8.61
12B	185	460	2.38	12	36.36	11.09
30B	1,400	5,020	26.0	29	68.02	52.21
48X	4,500	21,030	108.9	48	103.99	134.0
48Y	5,200	27,560	142.7	48	136.27	167.8

^aTable values are based on Ref. 7.

Table 4. Estimated Conditions for Failure of UF₆ Cylinders Exposed to Fire

Cylinder Characteristics			Hydrostatic Testing Results ^a			Estimated Fire Failure Conditions		
Type	Material	Wall thickness, in.	Failure mode	Failure pressure, psia	Volume increase, %	Failure pressure, psia	Volume increase, %	Final UF ₆ temperature, °F
5A	Monel	1/4				1900	20	434
8A	Monel	3/16				900	20	400
12B	Monel	1/4	Hoop stress	2265	53	800	20	396
30B	A516 steel	1/2	Hoop stress	2315	30	800	10	367
48X ^b	A285 steel	5/8	Brittle frac	1285	6.3	625	5	340
48Y	A516 steel	5/8	Brittle frac	1780	6.3	625	5	340

^aSee Refs. 8 and 9.

^bHydrostatic test results are from testing of a 48A cylinder.

conditions were estimated using UF₆ enthalpy correlations and a heat capacity for steel of 0.12 Btu/lb.°F.(13) It is conservatively assumed that the final cylinder wall temperature is equal to the final UF₆ temperature.

HEAT TRANSFER ANALYSIS

The starting point for evaluating the radiative heat flux from the fire to the cylinder is

$$Q = A_1 F_{12} \sigma (T_1^4 - T_2^4) / \left(\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} - 1 \right). \quad (2)$$

It is then assumed that the cylinder temperature is negligible relative to that of the fire. Noting that A₁ F₁₂ equals A₂ F₂₁ and assuming that the emissivity of the fire, ϵ_1 , is 1, the following equation for the radiant heat flux to the surface of the cylinder is obtained:

$$q_2 = 0.173 \times 10^{-8} F_{21} \epsilon_2 T_1^4. \quad (3)$$

For a cylinder totally immersed in a fire, F₂₁ = 1; for a cylinder external to a fire, the view factor from the effective surface of the cylinder to the fire, F₂₁, can be approximated based on the surfaces illustrated on Fig. 1. While the view factor correlation utilized in the approximation is itself rigorous,(14) the effective geometry shown on Fig. 1 is only an approximation; the illustrated geometry is expected to become more reasonable as the separation distance between the fire and the cylinder increases. Reported values for the emissivity of the cylinder, ϵ_2 , range from 0.3 or less for iron and steel to 0.95 for various paints and soot.(15)

The time to rupture for a cylinder exposed to fire is approximated by

$$\tau = 60 E/q_2 A_2. \quad (4)$$

Two cases are subsequently considered. The first assumes total immersion of the cylinder in the fire. The second assumes that the cylinder is outside the fire.

Case 1: A Cylinder Immersed in a Fire

It is assumed that the surface of a cylinder totally immersed in a fire rapidly blackens from soot; thus, it is reasonable to set the cylinder emissivity, ϵ_2 , equal to 0.95. Also, F₂₁ = 1.0 and

A₂ is the total surface area of the cylinder. The radiative heat flux from the fire to the cylinder is calculated by Eq. 2, then the time to cylinder rupture is estimated from Eq. 3. Estimated time intervals to rupture are given on Fig. 2 for a range of initial conditions and a flame temperature of 1475°F; specific results assuming solid UF₆ initially at 70°F are tabulated in Table 5. A multiplication factor to obtain the time to rupture at other flame temperatures is given on Fig. 3. For example, a 48X cylinder that is estimated to rupture in 27.3 min at a flame

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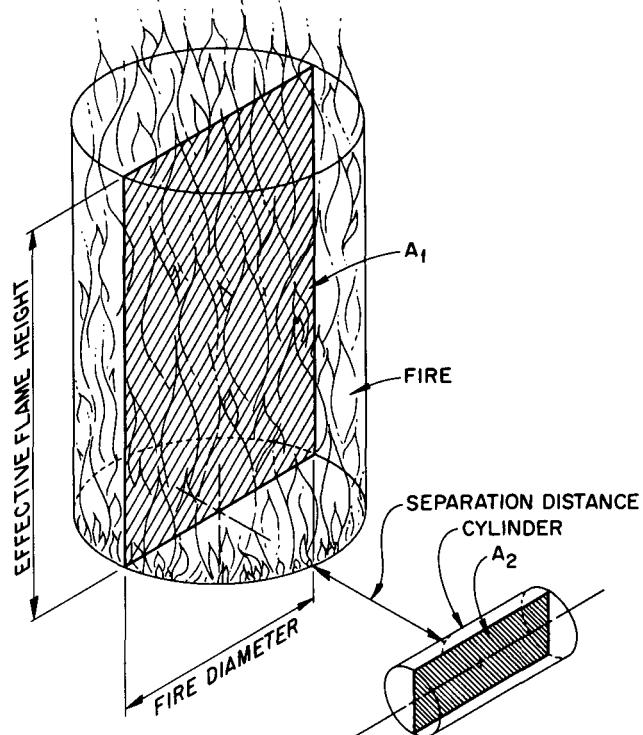


Fig. 1. Geometry for evaluating view factors between a fire and a cylinder.

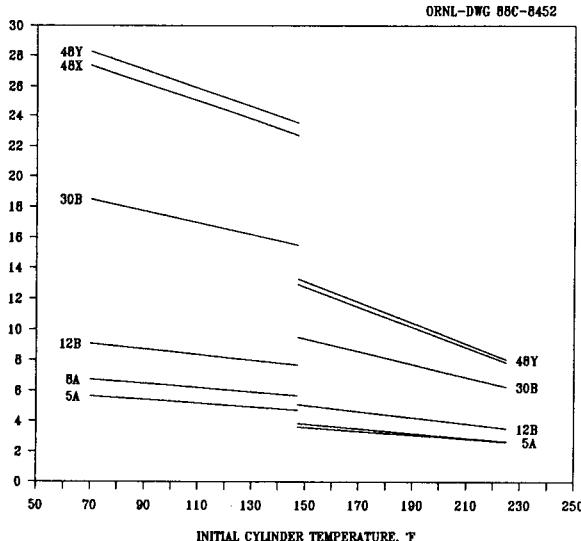


Fig. 2. Estimated time interval to cylinder rupture based on a flame temperature of 1475°F.

temperature of 1475°F would rupture at 12.3 min based on a 1900°F flame which yields a multiplication factor of 0.45.

Case 2: A Cylinder Adjacent to a Fire

For this second case, fires of several sizes were considered. Fire diameters at the ground surface of 10, 20, and 50 ft were selected, and effective flame heights twice the fire diameter were assumed based on the work of Mudan.(16) [Greater height to diameter ratios could have been assumed; but, since the fire is approximated as a right-circular cylinder (see Fig. 1) rather than as a cone, a ratio of 2 was considered a compromise.] Figure 4 summarizes view factors, F_{21} , from the cylinder to the fire; the view factors are not a strong function of cylinder size when separation distances exceed about 10 ft. A surface area multiplier, which is the ratio of the effective surface area (length x diameter) to the total surface area (see Table 4), is given in Table 6. For a cylinder

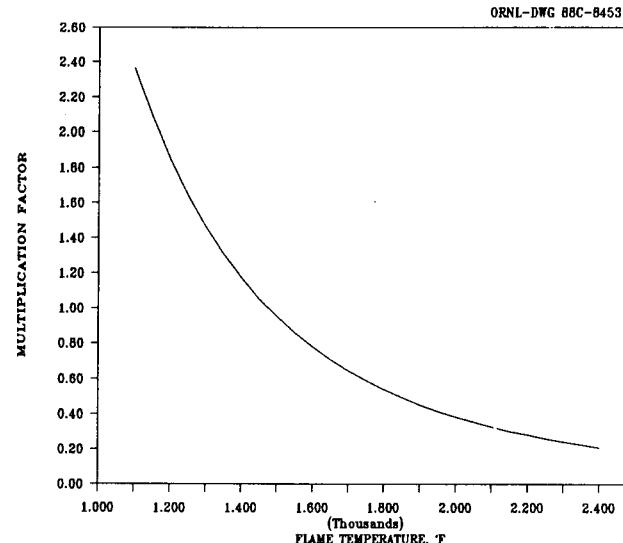


Fig. 3. Multiplication factor for adjusting the results given in Fig. 2 to temperatures other than 1475°F.

adjacent to a fire, its emissivity, ϵ_2 , can range from less than 0.3 up to 0.95, depending on the surface finish, as noted earlier.

To estimate the time to rupture for a cylinder adjacent to a fire multiply the time to rupture for a cylinder immersed in a fire (Fig. 2 or Table 5) by the flame temperature multiplication factor (Fig. 3) and the surface area multiplier (Table 6) and divide by the view factor (Fig. 4). If an emissivity other than 0.95 is assumed for the cylinder, multiply the result by 0.95 and divide by the assumed emissivity. For example, a 12B cylinder initially at 70°F will rupture in about 1 h when exposed to a 20-ft diam, 1900°F fire at a distance of 10 ft (i.e., $9.1 \times 0.45 \times 3.66 + 0.24 = 62$ min). Table 7 indicates time interval ranges needed to reach rupture conditions for a range of fire conditions.

RELATED STUDIES

In October 1965, cylinders containing from 5 to 250 lb of UF₆ were exposed to fire in a series of tests conducted at the Oak Ridge Gaseous Diffusion Plant (ORGDP). (17) These tests were conducted "to determine if the cylinders would hydrostatically or explosively rupture [and] the time available for fire fighting before either incident occurred." The cylinders were mounted where they would be completely within the fire. A summary of the tests is given in Table 8. During Test V, the cylinder wall temperature approached about 1000°F and UF₆ temperatures within the cylinder varied between 330 and 440°F at the instant the cylinder explosively ruptured. Mallett concluded that the tests "confirmed that [an] UF₆ cylinder rupture of explosive force is possible and that it can occur within a time sufficiently short as to possibly preclude fire fighting unless initiated very promptly. The explosions noted cannot be considered any more severe or hazardous than those due to other chemical or gas explosions. The amount of water blown from the tank by the force of

Table 5. Estimated Time Interval to Cylinder Rupture

UF ₆ phase	Solid	
Cylinder temperature	70°F	
Flame temperature	1475°F	
Heat flux	23,000 Btu/hr·ft ²	
Cylinder type	Total heat requirements, Btu	Time to rupture, min
5A	6,400	5.7
8A	21,900	6.8
12B	38,000	9.1
30B	364,000	18.5
48X	1,400,000	27.3
48Y	1,810,000	28.2

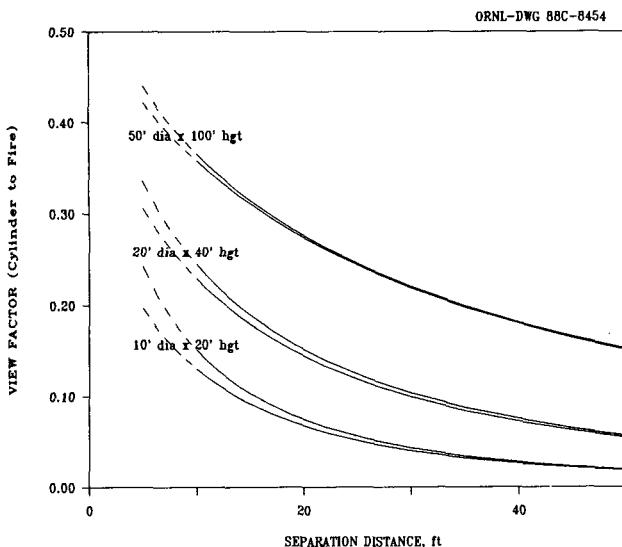


Fig. 4. View factors as a function of separation distance between fire and cylinder and fire size.

the explosion contributed largely to the fireball formation, a cause which, in most transportation accidents, is unlikely to be so available." Valve failures precluded explosions in Test I and IV.

Duret and Bonnard described the results of experimental and modeling efforts which included consideration of internal heat transfer in an UF₆ cylinder exposed to fire.(18) In a direct comparison with Mallett's results (which would be Tests III and V), they estimated a time to cylinder rupture of 8 min 40 s assuming a fire temperature of 800°C (1472°F); rupture would occur at an UF₆ temperature of 160°C (320°F). In their analysis, the cylinder wall temperature approached 600°C (1112°F) at the end of 6 min. Predicted failure durations for 30B and 48Y cylinders exposed to 800°C and 900°C fires were also presented (see Table 9).

UNCERTAINTIES IN THE ANALYSES

Direct comparison of time estimates to cylinder rupture based on the approach described herein (see Table 5) to the results of the ORGDTP tests (see Table 8) shows a conservative estimate of that time. Estimated times were 5.7 and 6.8 min for 5A and 8A cylinders, respectively, assuming a fire temperature of 1475°F, while Mallett's data indicated actual rupture occurred at 8 min for a 5A-sized cylinder and at 8.5 and 10.5 min for two 8A-sized cylinders. A number of conservative assumptions were made in these analyses; a nonconservative assumption is offset by the conservative assumptions. The various assumptions and their impacts--both in general, as well as on the comparison between calculations and experiment--are discussed in the following paragraphs.

Cylinder wall temperature. It has been assumed for these analyses that the cylinder wall temperature will have a negligible impact on the heat flux. However, Mallett's data, as well as the modeling of Duret and Bonnard, indicate that wall temperatures

Table 6. Surface Area Multipliers for Case 2

Cylinder type	Multiplier
5A	3.46
8A	3.42
12B	3.66
30B	3.81
48X	3.87
48Y	3.69

Table 7. Range of Time Intervals to Rupture for a Cylinder Adjacent to a Fire, min^a

Cylinder type	Separation distance (ft)		
	10	20	40
<u>Fire temperature: 1900°F</u>			
<u>Cylinder emissivity: 0.95</u>			
5A	16 - 57	20 - 116	31 - *
8A	16 - 69	21 - *	32 - *
12B	23 - 98	30 - *	46 - *
30B	44 - *	58 - *	89 - *
48X	62 - *	83 - *	*
48Y	62 - *	82 - *	*
<u>Fire temperature: 1475°F</u>			
<u>Cylinder emissivity: 0.95</u>			
5A	34 - *	45 - *	67 - *
8A	36 - *	47 - *	71 - *
12B	51 - *	67 - *	101 - *
30B	98 - *	*	*
48X	*	*	*
48Y	*	*	*
<u>Fire temperature: 1900°F</u>			
<u>Cylinder emissivity: 0.30</u>			
5A	49 - *	64 - *	97 - *
8A	52 - *	67 - *	102 - *
12B	73 - *	95 - *	*
30B	*	*	*
48X	*	*	*
48Y	*	*	*
<u>Fire temperature: 1475°F</u>			
<u>Cylinder emissivity: 0.30</u>			
5A	108 - *	*	*
8A	114 - *	*	*
12B	*	*	*
30B	*	*	*
48X	*	*	*
48Y	*	*	*

^aThe first number in each range corresponds to an initial condition of liquid UF₆ at the triple point (147.3°F) exposed to a 50-ft diameter fire; the second number corresponds to solid UF₆ at 70°F and a 10 ft fire. An asterisk, *, indicates a time greater than 2 h.

Table 8. Summary of ORGDP Fire Tests

Test	I	II	III	IV	V
<u>Cylinder Data</u>					
Diameter, in.	3.5	5	8	5	8
Length, in.	7.5	30	48	30	48
Material	Monel	Monel	Nickel	Monel	Nickel
UF ₆ mass, lb	5	55	248.9	53.04	245
<u>Failure Data</u>					
Mode	Valve failure	Explosion	Explosion	Valve failure	Explosion
Time, min	a	10	10.5	b	8.5

a. Two cylinders were tested simultaneously with valve failures occurring at 4 min and 6 min. The first failure occurred when teflon seals melted; the second when silver solder melted.

b. The two cylinder valves failed at 8 min and 9 min. The release was complete in 10 min.

exceeding 1000°F can occur. The reduction in heat flux resulting from the various wall temperatures is shown in Table 10. A further increase in the time to cylinder rupture would result from the heat capacity of the steel due to the additional temperature rise. Further analysis taking into account the complex phenomena of heat transfer within the cylinder is required to estimate cylinder wall temperatures.

UF₆ Enthalpy. The enthalpy of the compressed UF₆ at the point of rupture has been estimated from a correlation for saturated liquid enthalpy at lesser temperatures. This correlation is expected to underestimate the saturated enthalpy at higher temperatures. Accounting for the effects of compression, and improving the enthalpy correlation for higher temperatures, would increase the final enthalpy and, hence, the time to rupture.

Emissivity. In the analysis of a cylinder immersed in a fire, an emissivity of 1 was used for the fire. This assumption appears reasonable for large fires. However, relative to the argument of Buck and Belason, a fire emissivity less than 1 might be appropriate, based on the relative size of the fire and cylinders, for estimating the time relative to Mallett's data. Cylinder emissivity could be less than 0.95 which was chosen as an upper limit likely to be obtained in a fire environment. Lesser emissivities would increase the estimated time to rupture.

Table 9. Time to Failure for Cylinders Exposed to Fire (Estimates by Duret and Bonnard)

Cylinder type	Fire temperature, °C	Time to failure, min
30B	800	35
30B	900	28
48Y	800	61
48Y	900	47

Convective heat transfer. Convective heat transfer accounts for about 10% of the total heat flux in a fire environment. Inclusion of the convective component would decrease the time required to heat a cylinder to the point of rupture. Neglect of the convective flux is offset by the other assumptions already discussed.

Cylinder radiation and convection to environment

The cooling effects of radiation and convection from the cylinder to the environment for cylinders adjacent to a fire were not considered. Inclusion of such effects would increase the predicted time to rupture for cylinders not totally immersed in a fire.

CONCLUSIONS

The estimated time intervals to rupture for UF₆-filled cylinders exposed to fire should be considered preliminary, conservative estimates. Resolution of the various uncertainties discussed above should increase the estimated time intervals. The data of Mallett indicate that increased estimates are plausible. Consideration of cylinder expansion prior to rupture significantly impacts the time to rupture.

The estimated time intervals given on Fig. 2 and in Table 5 indicate that all sizes of cylinders may rupture within 30 min when totally immersed in a fire, although resolution of the uncertainties may

Table 10. Reduction in Radiant Heat Flux, %, Due to Cylinder Wall Temperature

Wall temperature, °F	Flame temperature	
	1475°F	1900°F
300	2.4	1.1
600	9.0	4.1
1000	32.	15.

increase time estimates for the 48X and 48Y cylinders beyond 30 min. For cylinders adjacent to fires, rupture of large cylinders appears much less likely. Test results show that valve failure may occasionally preclude cylinder rupture.

When a cylinder fails in a fire, the release of UF₆ into the fire will add energy to the fire.

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FLUORINE OVERPRESSURIZATION IN VHE (FIVE-INCH) CYLINDERS

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ABSTRACT

Uranium hexafluoride (UF_6) is thermodynamically a very stable compound under normal conditions. However, UF_6 is subjected to self-irradiation (due to natural alpha decay) and, as a result, can decompose to UF_x and fluorine (F_2). Very highly enriched UF_6 (VHE) contains significant quantities (1-2 percent) of the isotope U-234 which has one of the highest specific activities of the more commonly encountered uranium isotopes. The intense self-irradiation from the U-234 alpha particles results in a slow and spontaneous decomposition of UF_6 . Operating experience with cylinders of VHE after extended storage has demonstrated the existence of non-volatile deposits (heels) and overpressurization due to F_2 . Studies on the estimated rate of UF_6 decomposition and the results of examinations of cylinder heels will be discussed.

INTRODUCTION

The storage and transfer of very highly enriched top product (VHE) at the Portsmouth Gaseous Diffusion Plant are accomplished in nuclearly safe 5-inch cylinders with minimum spacing of 2-feet. During the more than 30 years of VHE production, small, but significant and variable, amounts of non-volatile deposits have been encountered in cylinders and sampling lines during VHE transfer operations. Deposit-laden systems may also exhibit abnormal elevation of pressure especially after UF_6 has been pre-heated in preparation for liquid UF_6 sampling. The handling procedures in these instances have been modified to address cylinder overpressurization, plugging of the liquid sampling system, and cylinder heels caused by non-volatile material.

The recent increased emphasis on uranium accountability has demanded that greater priority be placed on quality sampling and accurate uranium measurements. These operations can be adversely affected by heterogeneous samples caused by deposits in UF_6 -containing systems. Of even greater concern, however, is the overpressuring of UF_6 cylinders, which presents a potential safety issue. This paper describes the results of studies to identify the basic causes and magnitude of this overpressurization of 5-inch cylinders and

the results of examinations of cylinder heels. Results of earlier investigations conducted by Bernhardt et al at the Oak Ridge Gaseous Diffusion Plant(1) have been used to interpret the observed solids deposition and cylinder overpressurization.

BASIC OBSERVATIONS OF OVERPRESSURIZATION IN 5-INCH CYLINDERS

Instances of abnormal pressure buildup in VHE cylinders have been encountered at numerous times over the years during pre-sampling heating at the High Assay Sampling Area (HASA). The following general observations are associated with cylinder overpressurization:

1. There is a correlation between cylinder overpressure and the time the VHE cylinder has been in storage. Newly-filled cylinders, for example, do not display abnormally elevated pressures. The magnitude of the overpressuring has been as high as three atmospheres above the expected vapor pressure of UF_6 at the prevailing temperature.
2. Deposits of non-volatile solids are frequently found in the lines of the liquid UF_6 sampling systems. In many cases these solids cause plugging problems which require special procedures (e.g., filtration) to obtain the required UF_6 samples for analytical measurements.
3. The VHE cylinders may contain non-volatile heels in quantities varying from a few hundred grams to greater than a kilogram.

In addition, samples of gas obtained from overpressurized cylinders have been identified as fluorine (F_2) in substantial concentrations (greater than 80 mole percent), and samples of cylinder heel material have been identified as uranium pentafluoride (UF_5). The UF_5 has the beta-form crystalline structure, which is the form expected when UF_6 is reduced at temperatures below 300°F. There have also been efforts to identify the solids within liquid sampling equipment but their amorphous nature generally precluded structural determination. In one instance, however, a sample removed from a liquid sampling block was identified as UF_5 . Direct wet chemical analysis of other deposits confirmed the high uranium con-

tent. It is likely that these materials are also UF_5 as in the case of the cylinder heels.

Of the number of possible causes of the above observations, the decomposition of UF_6 due to self-irradiation is considered the most likely explanation of both the overpressurization and heel formation encountered with VHE cylinders. A detailed look into a mechanism, which has been largely disregarded in UF_6 handling operations, is provided below.

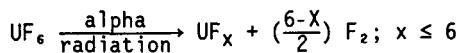
DECOMPOSITION DUE TO ALPHA IRRADIATION

Past studies on the alpha irradiation of UF_6 , using radon as the alpha source, form the basis of much of the discussion presented here(1). The results of these studies applied to VHE product characteristics, present a convincing case that self-irradiation of UF_6 by alpha particles results in its dissociation and formation of UF_x and F_2 . The extent of this decomposition is largely dependent on the U-234 content and time. UF_6 at VHE assays contains significant levels of U-234, which has a considerably higher specific alpha activity than U-235, U-236 or U-238 (Table 1).

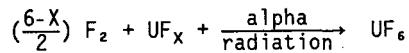
TABLE 1 SPECIFIC ACTIVITIES OF U-ISOTOPES
(disintegrations/sec/gram)

U-234	1.54×10^8
U-235	5.34×10^4
U-236	1.58×10^6
U-238	8.34×10^3

When an alpha particle strikes a UF_6 molecule, approximately 1.37×10^8 ion pairs are formed over its path length. (This figure is based on a gas phase determination and is assumed to also be the approximate value for the solid phase.) VHE product decomposition occurs as a result of self-irradiation by alpha particles according to the following equation:



Although theoretically the solid UF_x compound may have any stoichiometry intermediate between UF_6 and several subfluorides (UF_5 , UF_4 , etc), only beta uranium pentafluoride has been identified as the primary solid uranium fluoride in cylinder heels. The fluorine produced by this reaction would be expected to build up slowly in a sealed container at a rate determined by several factors. It might be expected that over a sufficiently long period of time, all the UF_6 would be eventually reduced to solid UF_x . However, a reverse reaction, also induced by alpha particles, can oppose the decomposition as follows:



The fluorination of UF_x compounds with fluorine normally occurs at a significant rate only at high temperatures. UF_4 , for example, is not rapidly converted to UF_6 by F_2 until temperatures above 400°F are present(2). However, UF_x deposits (previously formed by radon alpha decomposition of UF_6) can be converted at measurable rates back to UF_6 in the presence of F_2 at room temperature under the influence of alpha radiation(1). Under

self-irradiation exposure, the decomposition of VHE UF_6 (in a sealed system where the F_2 cannot escape) would be expected to reach an equilibrium at which point the UF_x deposit formation rate equals the conversion rate back to gaseous UF_6 . The rate of the reverse reaction is a function of the available surface area of the UF_x deposit as well as the F_2 activity for a given radiation environment.

DISCUSSION

Calculations were made to estimate the quantity of fluorine and UF_5 produced based on this decomposition model. The method employed to estimate an upper bound for the degree of alpha-induced decomposition assumed (1) no reverse reaction, (2) no container wall effects, (3) every ion pair formed leads to the decomposition of one UF_6 molecule. The specific activity and the nominal value (1.37×10^8) for the number of ion-pairs created per alpha particle were used to calculate the quantity of UF_5 and F_2 produced for a given time period.

The specific activity (SA) per gram of VHE UF_6 of isotopic composition given in Table 2 was calculated using the equation:

$$SA = \sum \frac{\lambda_i A_i W_i}{M_i} \text{ (dps/g)}$$

where: λ_i = decay constant of isotope i
= $0.693/t_{1/2}$, where $t_{1/2}$ = half life
in seconds
 A = number of molecules/
mole = 6.023×10^{23}
 W_i = isotopic weight fraction
 M_i = molecular weight, grams/mole

TABLE 2 ASSUMED ISOTOPIC COMPOSITION OF UF_6 FOR CALCULATION OF DECOMPOSITION RATE

U-234	1.20%
U-235	97.35%
U-236	0.0%
U-238	1.45%

The results indicate that decomposition of UF_6 to UF_x and F_2 , due to self-irradiation, would be on the order of 0.48% by weight per year. The estimated quantity of fluorine generated in one year for 25 Kg of UF_6 (fill limit for VHE cylinders) is 6.5 grams. With a free space of 1 liter, this quantity of F_2 would be expected to produce a pressure in excess of 60 psia at room temperature, neglecting the solubility of F_2 in solid UF_6 and corrosion losses. The quantity of UF_5 heel produced is calculated to be 113 grams.

In addition to U-234, the presence of other radio-nuclides of high specific activity, and at sufficiently high concentrations, would be expected to contribute to UF_6 decomposition. Decomposition due to the uranium isotopes U-232 and U-236 is not significant since the concentration of U-236 has historically been below 0.02 weight percent in VHE material, and no U-232 has been detected in VHE at the analytical limits of sensitivity (2 parts per billion, U-basis). The only non-uranium radioactive contaminant encountered in VHE product that has the potential for radiation-induced UF_6

decomposition is technetium-99, which is a weak beta emitter present in trace amounts in recycled uranium from reactors. While the specific activity of technetium-99 is nearly three times that of U-234, the energy of its beta particle is considerably less than that for the alpha particles from uranium isotopes. Additionally, the concentrations of Tc-99 in VHE product are typically four to five orders of magnitude below those of U-234; therefore, no significant effect from the presence of Tc-99 is expected.

CONCLUSIONS

The calculated pressures of F_2 created as a result of the dissociation of UF_6 due to self-irradiation correlate with pressures actually observed and therefore supports the model for the production of cylinder overpressurization and heel formation. The solid in cylinder heels and sampling systems has been identified as beta UF_5 , while the gas contributing to the cylinder overpressure is F_2 .

This model is also supported by observations showing that the amount of F_2 overpressure is a function of storage time. G. B. Binstock of E. I. DuPont (Savannah River Plant) has also observed that the quantity of VHE cylinder heel varies directly with cylinder storage time(3). There is a lack of data, however, on whether the relationship is linear or whether it begins to plateau due to a reverse (fluorination) reaction. This is considered an important unknown aspect of the problem since it would determine to what extent

the pressure buildup is self limiting and might provide approaches to minimizing or managing the problem more efficiently.

Overpressurization in 5-inch (VHE) cylinders and the formation of non-volatile heel material requires special attention during handling and transfer of VHE product. In addition to the safety aspects of the F_2 pressure buildup in heated cylinders, there is interest in heel formation from the standpoint of improved sampling and uranium accountability. The operational approach at Portsmouth has been multi-faceted and involves special procedures for handling the overpressurization and heel problems, while maintaining the storage time as short as possible.

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THE TOXIC AND RADIOPHYSICAL RISK

EQUIVALENCE APPROACH IN UF_6 TRANSPORT

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ABSTRACT

After a brief description of the current situation concerning safety in transport of UF_6 and of the work by the IAEA on developing new regulations, we go on to discuss the equivalence of the radioactive and chemical risks in UF_6 transport regulations.

As the concept of low specific activity appears to be ill-suited for a toxic gas, we propose a quantity of material limit designated T_2 (equivalent to A_2 for radioactive substances) for packagings unable to withstand accident conditions equal to those laid down by the IAEA (9 m drop, 800°C fire environment for 30 minutes).

It is proposed that this limit be chosen for the amount of release acceptable after IAEA tests for packagings with a greater capacity than T_2 kilograms.

Different possible scenarios are described, with fire assumed to be the most severe toxic risk situation.

The risk equivalence approach leads to requiring that the packaging be capable of resisting a 800°C fire for 30 minutes without the amount released exceeding T_2 . The problem of demonstrating the behaviour of the shell and the openings (especially the valve) is raised in this context.

1. INTRODUCTION

The experience which has been acquired over the last thirty years in the transport of uranium hexafluoride, during which some tens of thousands of tonnes of natural, depleted and enriched uranium hexafluoride were carried, shows that the safety level of the packagings now universally used is substantial. None of the few accidents which have occurred have had consequences for the public or the environment. Nevertheless, this must be considered in the light of the fact that none of the accidents which took place subjected the packages to severe conditions, such as those adopted by the IAEA, under which the chemical risk of the uranium hexafluoride could represent a major problem.

It is for this reason that, after the Mont Louis accident, the IAEA began work on developing regulations covering both the radiochemical and chemical risks of uranium hexafluoride. Although the project presented in technical document TEC-DOC 423 [1], now being discussed may have the advantage of filling a gap as concerns allowance for the regulatory level of the chemical risk for high-capacity packages, which had not previously been taken into consideration [2], it is nevertheless not fully satisfactory. The report acknowledges this shortcoming as it emphasizes that the logical approach is to guarantee an equivalent radiological and chemical risk but that such an approach needs to be given careful consideration, being liable to call into question all the existing regulations in view of the different principles on which regulations for the transport of radioactive substances and other dangerous substances are based. The purpose of this paper is to present a certain number of views concerning this problem which are in agreement with others previously expressed concerning the same subject, particularly by M. Biaggio [3] and by the CEA [4].

In this first part, no reference is made to the existing packagings, which certainly offer a level of safety equivalent if not higher than that generally applied in protective measures against similar chemical risks, although the safety level of these packagings has never, to our knowledge, been evaluated.

The IAEA regulations [5] for radioactive substances are essentially concerned with obtaining an equal level of safety whatever the substances, their quantities and the performance of their packaging, under accident conditions which correspond to a high percentage of accidents, and which is defined on the basis of regulatory tests; this safety level has been set extremely high as the requirement is to limit the radiological effects to the equivalent of the annual dose allowed for workers (ALI). It will be noted that this level offers an ample safety margin with regard to the lethal risk, being of the order of more than 100.

Using this objective, the IAEA regulations impose performance criteria, leaving the designer entirely free to choose his own solutions for the design of the packaging.

As concerns allowance for the chemical risk, a similar approach can be adopted. This means that the first step is to set a safety level considered to be acceptable under the same accident conditions as those applied by the IAEA, i.e. those corresponding to the "B tests".

2. CHEMICAL AND RADIOPHYSICAL RISKS ASSOCIATED WITH URANIUM HEXAFLUORIDE

Setting an acceptable limit for the chemical risk.

Uranium hexafluoride has physical properties which set it apart from the immense majority of radioactive substances transported. It is solid under pressure and at ambient temperature, becoming a liquid at relatively low temperature and pressure, and a gas at normal pressure at 56°C. It is transported in solid form but as it rapidly reacts with compounds containing hydrogen, if there is the slightest leak in its packaging, it reacts with the water always present in the atmosphere to form aerosols of UO_2F_2 and gaseous hydrofluoric acid. As these two products are strongly hygroscopic, they themselves become hydrated. The presence of UO_2F_2 is easily detected by the presence of a cloud of white smoke. One finds therefore in the environment, whenever there is a leak in transport, new products which require consideration in evaluating consequences: UF_6 in gaseous form which has not reacted with atmospheric humidity, UO_2F_2 in the form of an aerosol of fine particles and HF in gaseous form.

The consequences in the event of accidental release are then associated, on the one hand, with the effective equivalent dose resulting from the inhalation of uranium (radiological risk) and, on the other hand, the toxic effect of uranium (from the UO_2F_2) and the toxic effect of hydrofluoric acid (from the HF and the UO_2F_2).

The radiological risk increases with the degree of enrichment; UO_2F_2 being one of the most soluble uranium compounds, the effect is mainly concentrated in the kidneys. Hydrogen fluoride is extremely corrosive and in the event of heavy exposure there are serious results for the eyes, the skin and the respiratory system.

The effects of the direct inhaling of UF_6 are the combined effects of UO_2F_2 and HF, as hydrolysis occurs in the lungs.

The toxic risk of the uranium is thus present at the same time as the toxic risk of the HF. In this paper, we shall only consider the toxic effect of the HF. Indeed, the respective lethal levels of UO_2F_2 (200 mg) and HF (15 mg), the equivalent level of discomfort for UO_2F_2 (25 mg) and HF (5 mg), for a short duration exposure of the order of half an hour, are of the same order for the release of a given quantity of UF_6 .

The toxic risk of HF is represented by the curves in Figure 1 [6] which have been plotted with allowance made for the different harmfulness levels. Unlike the radiological and toxic risk of the uranium, the toxic risk of HF depends more on the concentration than the duration of exposure. Thus, between the discomfort level and the lethal level, the duration of exposure is multiplied by 200, whereas the concentration is only multiplied by 10.

For the chemical risk equivalence, we propose to consider the discomfort level acceptable, which means that an individual will not be significantly affected under the conditions of exposure to the products present immediately after a release of UF_6 . It should be noted that this level is already considerably less severe than that accepted for the radiological risk in transport, as it results in a safety margin with respect to the lethal risk of only a factor of 10 instead of 100.

3. COMPARED RISKS UNDER NORMAL AND ACCIDENT CONDITIONS

Normal conditions (Figure 2)

The average daily limits generally accepted for workers are set at 1 ALI/250 (a debatable mean value)* for the radiological risk, 2.5 mg of uranium (3.7 mg of UF_6) and 2 mg of HF (6 mg of UF_6) for the toxic risk. The toxic risk of the uranium is, under these conditions, in all cases greater than the toxic risk of the HF, and the radiological risk becomes greater at more than 3% enrichment, while remaining extremely slight as it only corresponds to 1 ALI/250. Although this data must be taken into consideration for the safety of installations, the same does not apply to transport as the IAEA regulations require the packagings to be hermetic under normal operating conditions and the transport operators are not considered to be exposed to this risk for as long.

Accident conditions (Figure 3)

The principal problem involved in the transport of toxic substances is the behaviour of packages under accident conditions.

The regulations covering the transport of radioactive substances require that ALI is not exceeded in accident conditions, the hypotheses applied being exposure for a short duration of the order of 30 minutes.

The corresponding quantities of HF which can be released lead, even for the greatest levels of enrichment, to unacceptable values from the chemical point of view, as the lethal risk is reached with enrichment of about 20% and a higher level than for discomfort is attained with 90% enrichment. The same applies for the toxic risk of the uranium. Indeed, the quantity corresponding to 1 ALI is greater than the quantity of uranium which, inhaled at the same time, would be liable to cause lethal kidney damage by chemical toxicity (about 200 mg) up to 15% enrichment. Up to 90% enrichment, this quantity is greater than the quantity which it would be possible to inhale daily (2.3 mg) without permanently damaging the kidneys.

The toxic risk exceeds the radioactive risk in transport accident conditions and the radiological criterion corresponding to 1 ALI does not allow the proposed equivalent toxic risk to be satisfied at the same time, which corresponds to a discomfort level of the order of 5 mg inhaled in half an hour of exposure.

Using the same hypotheses as those adopted by the IAEA to define packagings unable to withstand accidents, an attempt can be made to evaluate the maximum quantity of UF_6 which is possible to transport under these conditions. Considering the three plausible accident scenarios, in a standard hall (300 m³, 4 air changes per hour and presence for 1/2 hour):

- large breach at room temperature: quantity released 1%,
- large breach followed by fire: quantity released by sublimation or melting: 100%,
- uncontrolled fire: quantity released 100%.

*One may wonder about the validity of the use of the ALI (5.10⁴ Bq) for the soluble compounds of uranium such as UO_2F_2 which is calculated on purely radiological criteria.

The acceptable quantities are respectively of the order of 10 kg and about one hundred grammes. The fire case is the most severe, especially as the aerosol concentration rapidly reaches levels at which visibility is practically zero, making evacuation extremely difficult.

The fact that it is forbidden to store packagings of UF_6 in halls close to combustible materials should make it possible to make allowance only for accident scenarios involving rupture at room temperature.

This means that to cover the toxic risk, all packagings containing more than 10 kg of UF_6 must be able to withstand the accident conditions laid down by the IAEA with an allowable amount of leakage on completing the tests which can be established on the basis of the consequences of an accident outside, the occurrence of an accident of the severity of those provided for in the B tests being unlikely within a building. This results in an allowable leak rate of the order of 6 g/s for HF or 20 g/s for UF_6 , assuming discharge for 30 minutes at ground level under normal conditions, in order not to exceed the discomfort level at a distance of 100 m (Figure 4).

Can the checking of the degree of leaktightness be limited to a duration of half an hour? It is reminded that, for the radiological risk, the IAEA imposes a duration of one week as it assumes that intervention can last for one week. A duration of intervention of 30 minutes is certainly extremely optimistic. Indeed, this assumes that the emergency teams would have brought the accident under control within half an hour (which is scarcely credible in the case of a fire with a duration of half an hour), is that the population concerned would have been evacuated within this time. A plausible accident scenario is a fire with leakage of the valve initiated soon after the start of the fire and continuing for a certain length of time after the fire has stopped. We estimate at an additional 1/2 hour the time necessary to stop the leak by placing a wet cloth over the valve. The leakage being slight, the product will tend to solidify and rapidly form a plug by cooling as it passes through the valve.

These hypotheses lead to defining an allowable leak in a period of one hour from the start of the fire; allowable leak rates would then be 3 g/s of HF, i.e. 12 g/s of UF_6 .

4. THE CURRENT REGULATORY SYSTEM: APPLICATION OF IAEA REGULATIONS

From the strict application of the IAEA regulations, it appears that the problem covering the toxic risk is only present for less than 1% enrichment as once this limit is passed, the packages must meet the criteria for fissile packages. For UF_6 , this condition requires the packagings to remain hermetic after the accident tests, which is stipulated to meet the need to guarantee maintaining controlled moderation.

At lower than 1% enrichment, the toxic risk is properly prevented with our hypotheses only for transported quantities of less than about 10 kg, as the packagings required do not have to be capable of withstanding accidents.

It will be noticed that the recommendations put forward in the IAEA draft – TEC-DOC 423 currently under discussion, do not cover the toxic risk either, as the recommendations make no reference to design basis accidents, in particular to the case of packages of less than 450 litres which are precisely those for which the problem of being able to withstand fire is the most crucial.

5. IS THE RADIOPHYSICAL RISK OF UF_6 CORRECTLY COVERED BY THE IAEA REGULATIONS?

Allowance for the toxic risk leads to raising in the question, which may appear surprising, of the radiological risk and whether it is covered by the existing regulations at less than 1% enrichment. Indeed, UF_6 is considered to be a solid substance of low specific activity (TEC-DOC 423).

Is UF_6 really a solid? §208 of S.S. No. 6 rightly mentions that allowance should be made for the forming of other dangerous substances subsequent to reaction with the atmosphere in the event of rupture of the containment envelope due to an accident. Indeed, it is the physical state in which the substance is present when the packaging is faulty which is the one which requires consideration. However, as emphasized in this document, UF_6 is then no longer a solid, but has become, by dispersion, a gas on breaching of the envelope which then becomes, at a rate depending on the conditions of the accident, an aerosol of UO_2F_2 and gaseous HF.

Therefore, in our opinion, UF_6 should be considered to be a gas for regulatory purposes. It cannot therefore be placed in class LSA I. Can it be placed in class LSA II? Here we come to a delicate matter as the present regulations base the concept of substances of low specific activity on the axiom that it is highly improbable for an individual to inhale more than 10 mg of a substance. This value is controversial, even for solids, as there are examples of inhalation of uranium powder by miners which have reached quantities of a number of hundreds of milligrams [7], and is difficult to accept for gases. This means that the value of A_2 for a gas cannot be unlimited and that the problem of the value of A_2 is raised. The concept of A_2 being equal to 10^6 ALI is effectively based on the hypotheses that when a packaging of type A is destroyed, only between 1/100 and 1/1000 of its content is in dispersible form, and a fraction between 10^{-4} and 10^{-3} of this quantity is inhaled.

The authors of system Q [8] considered that this value of 10 mg could only be exceeded in extreme cases which should not need to be taken into account. Do not gases, by their physical nature, constitute one of these extreme cases?

In the case of UF_6 , depending on the accident scenarios (rupture at room temperature with a large or small breach, or rupture under fire conditions), the proportion of UF_6 transformed into gaseous UF_6 or into an aerosol of fine particles of UO_2F_2 may vary considerably, from values below one thousand to unity. The worst case would then lead to giving A_2 a value of 10^6 ALI instead of 10^6 ALI. Therefore, the maximum quantity of natural UF_6 (ALI equal to 5.10^4 Becquerels) which could be transported in a type A packaging would only be 20 kg instead of the 12 t regularly carried!

This quantity is nevertheless twice the allowable quantity for the toxic risk and the allowed quantity would be 30 times higher if the radiological and toxic risks were the same with respect to the lethal risk (safety factor of 10 with regard to the lethal risk). The result is that this does not change our proposal to provide resistant packagings for quantities greater than 10 kg, as it is the toxic risk which predominates.

It is a debatable matter as to the quantity that an individual can inhale in the form of a gas or aerosol in a transport accident of which the duration is always extremely brief. Can an individual inhale more than 1.2 g of uranium (ALI of natural uranium)? If it is accepted that a maximum quantity of some hundreds of milligrammes is possible, as this value is lower than the ALI, it can be accepted under these conditions that A_2 is unlimited and there is no radiological limit on the quantity which can be transported in a non-resistant packaging as permitted under the regulations. This question needs more clarification.

The case of reprocessed UF_6 less than 1% enriched requires particular attention. Once it is considered that UF_6 is a gas or even a dispersable LSA material, the current radiological regulations (1985) where $A_2 = 10^6$ ALI, impose a limit of 100 A_2 per vehicle. Thus, in view of the specifications for U 232 and impurities (transuranic elements and PF) of the UF_6 originating from reprocessed uranium, the quantity allowed per packaging for a non-resistant packaging would be less than a value of 12 tonnes.

6. HOW DOES THE UNIVERSALLY USED PACKAGING BEHAVE?

The packagings in use for more than thirty years correspond to extremely detailed standards [9] as concerns their design, and fabrication, inspection and maintenance specifications, which cover all degrees of enrichment. Although the behaviour criteria are imposed for fissile material at more than 1% enrichment, this is not the case at lower degrees of enrichment. Nevertheless, they have thermal and mechanical properties which must be taken into consideration in determining the consequences in accident conditions. Unfortunately, the performance levels have not been evaluated in a systematic manner, especially as concerns the thermal behaviour of the packaging and the resistance of the valves, which ought to be the subject of research programmes as emphasized in document TEC-DOC 423.

For less than 1% enrichment, current practice is to use type 48 Y high-capacity packagings, with a capacity of 12.5 t of UF_6 .

Certain mechanical and thermal evaluations have been carried out which show, pending confirmation, that the type 48 Y packaging has characteristics which seem to meet the TEC-DOC 423 requirements, at least as concerns the overall behaviour, the problem of the valve remaining whatever the case. Indeed, drop tests carried out in the USA 20 years ago on a type 48 G thin-wall packaging [10] have shown that this packaging can withstand a 9 m fall. Nevertheless, this result should be reviewed in the light of the accident at Portsmouth in 1978 when an identical packaging filled with liquid UF_6 developed a breach after a drop of a height of less than one metre onto a wooden block opposite the reinforcement. It is true

that this type of accident is not provided for by the IAEA! No fire tests have been carried out on this package in the presence of UF_6 , but only calculations of which the results offer a considerable margin of incertitude is difficult to quantify due to lack of understanding of the physical models. The calculations made by the CEA indicate rupture occurring after between 30 and 60 minutes [11] for a 800°C fire, by a hydraulic process.

Some consider that rupture would more probably be of an explosive nature (gas pressure). A test programme in progress, in Japan in particular, should make it possible to develop a universal physical model for the behaviour of UF_6 in the presence of fire [12].

As concerns the resistance of the valve, it is certain that there is no case where leaktightness can be guaranteed. The thermal calculations for a 800°C 1/2 hour fire [11] indicate that the valve would begin to leak 5 minutes after the start of the fire due to melting of the brazed joint applied to the thread, which takes place at 230°C. The leak rate depends on the position of the valve at the moment of the accident (up position: leakage in gaseous phase throughout the fire with, using our hypotheses, the liquid level only reaching the valve after 30 minutes, down position: two-phase leakage or gaseous phase leakage depending on the passage cross-section).

It may be considered that, in the event of a 9 m drop, the valve would be broken, and that there would be slight leakage by slow hydrolysis, UO_2F_2 , contributing to delaying the progress of the reaction. In any case, as rapid plugging is possible, this type of accident does not constitute a problem.

Let us consider the consequences of the following four accident cases.

1. Nine metre drop

The cylinder remains intact but the valve breaks, resulting in a slight leak by slow hydrolysis, the UO_2F_2 , contributing to delaying the reaction. In any case, as rapid plugging is possible, this type of accident does not constitute a problem.

2. 800°C fire for half an hour

The cylinder remains intact but the valve leaks. Slight leakage occurs at the thread of the valve due to melting of the brazed joint at 200°C. As this leak would be of low amplitude, it is assumed that it would be in the vapour phase (rapid vaporization of liquid UF_6 on passing through the hot valve) and that its rate would be directly proportional to the temperature of the UF_6 . The mean leak rate during the fire is estimated at 3 g/s per mm^2 of passage cross-section. Assuming a possible passage cross-section of about 10 mm^2 (half the area), the mean rate would be 30 g/s of UF_6 , ie a HF equivalence of 10 g/s. After the fire, the leak would continue at a decreasing rate and stop after 1/2 h according to our hypotheses.

For evaluation of the consequences, two phases require consideration:

- During the fire (duration 1/2 h, leak rate 10 g/s)

A turbulence and plume effect must be allowed for as the fire, involving a high-capacity packaging, is of a severe nature. Assuming the plume to be 25 m high under DNS conditions, no ill-effects need be feared (Figure 6).

- During the half hour following the fire

Release will occur at ground level. As in the preceding phase the plume effect had protected an area extending out to 500 m from the site of the accident, it is considered for the purposes of evaluation of consequences at 100 m that release begins when the fire is extinguished. Assuming the same leak rate, ie 10 g/s of HF (which is extremely conservative), for 1/2 h, the consequences are acceptable, being close to the level set for discomfort (Figure 4).

3. Nine metre drop with breaking of the cylinder followed by an 800°C 1/2h fire

The UF₆, which is solid at atmospheric pressure, will sublime during the fire. Dispersion of a large quantity of the substance will take place during the fire and continue for a number of hours after it if action is not taken. Assuming that action is rapidly taken and that the leak is stopped after one hour, roughly half the content will have escaped [14], ie 6 t of UF₆, which is equivalent to 1.5 t of HF. At a constant leak rate, this represents 2 kg of HF per second. Allowing for the plume effect, lethal effects will be observed at distances of 500 to 600 m from the site of the accident (Figure 7).

4. Uncontrolled fire

Should the cylinder fail to resist an 800°C fire for 1/2 h or should the fire last longer, the cylinder would eventually break and release its entire content virtually instantaneously, ie 12.5 t of UF₆ or 4.2 t of HF.

Lethal effects will be observed at distances of 500 to 1000 m (Figure 7).

7. CONCLUSION

The regulations concerning the transport of radioactive substances provide a high degree of safety which is equivalent for the toxic and radiological risks in the transport of more than 1% enriched UF6. This is not the case for less than 1% enriched UF₆ for which the chemical risk of the UF₆ is the main one. Two approaches then become possible to guarantee an acceptable level of safety. The first approach would be to apply the same constraints as for chemical products involving a similar type of danger, for example hydrofluoric acid or chlorine, i.e. to follow the UNO recommendations for class 8. This is the approach which has been adopted in developing TEC-DOC 423. The other approach would be to apply criteria equivalent to those used for the radiological risk.

It is this second approach which we recommend be developed in the long term. This approach is consistent with the one used by the IAEA to develop the regulations for the transport of radioactive substances, ie of first setting an objective then establishing criteria for reaching it, indeed appears to be the only one liable to guarantee a known level of safety as it is the one which is self-imposed, while leaving full scope for innovation as it is criteria which are laid down and not specifications.

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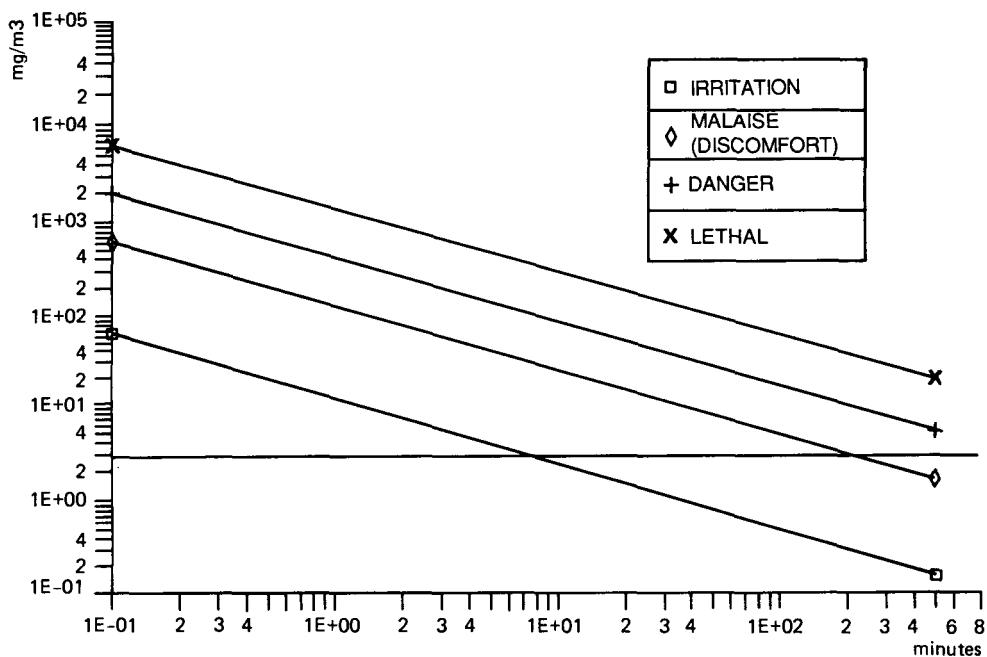


Figure 1
TOXICAL EFFECTS OF ANHYDROUS HF VERSUS CONCENTRATION AND TIME

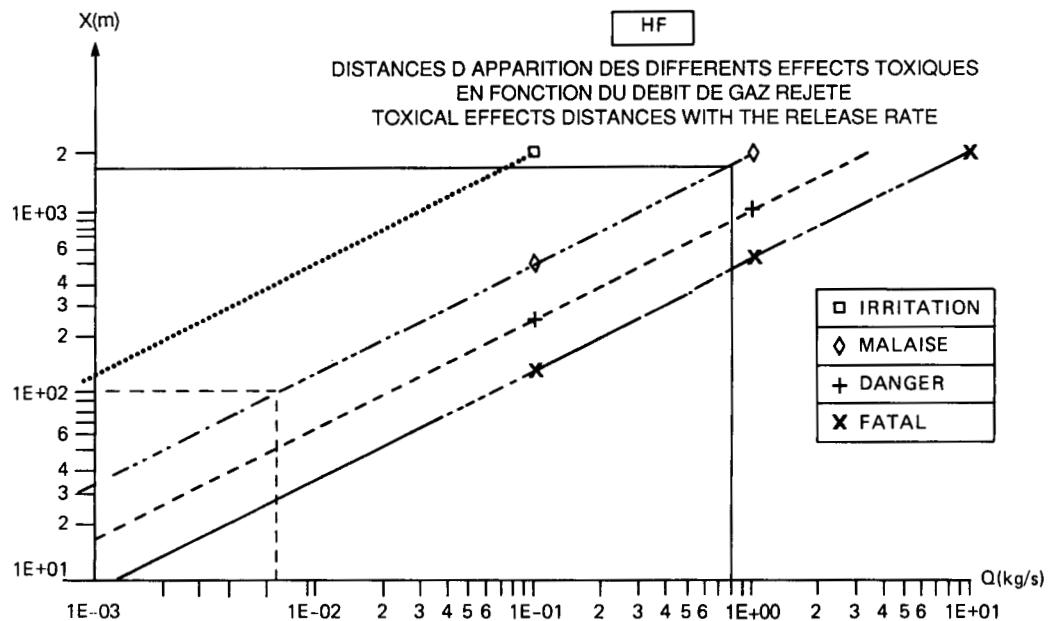
ENRICHMENT % U235	SPEC. ACTIVITY	EQUIVALENT MASS TO ALI/250	RADIOLOGICAL RISK	TOXICAL RISK U	TOXICAL RISK HF
%	Bq/g	mg	UF ₆ EQUIVALENT MASS TO mg		
NATUREL	2,610 ⁴	8	12	3,7	6
1%	2,810 ⁴	7	10.5	3,7	6
3%	5,10 ⁴	4	6	3,7	6
5%	10 ⁵	2	3	3,7	6
10%	1,810 ⁵	1.1	1.65	3,7	6
50%	9,310 ⁵	0.20	0.30	3,7	6
90%	2,210 ⁶	0.09	0.14	3,7	6

Figure 2

ENRICHMENT % U235	URANIUM MASS EQUIVALENT TO 1ALI	UF ₆ MASS	HF MASS
%	mg	mg	mg
NATUREL	1,900	2,800	950
1%	1,800	2,650	900
5%	500	750	250
10%	300	450	150
20%	145	220	75
50%	50	75	23
90%	20	30	9

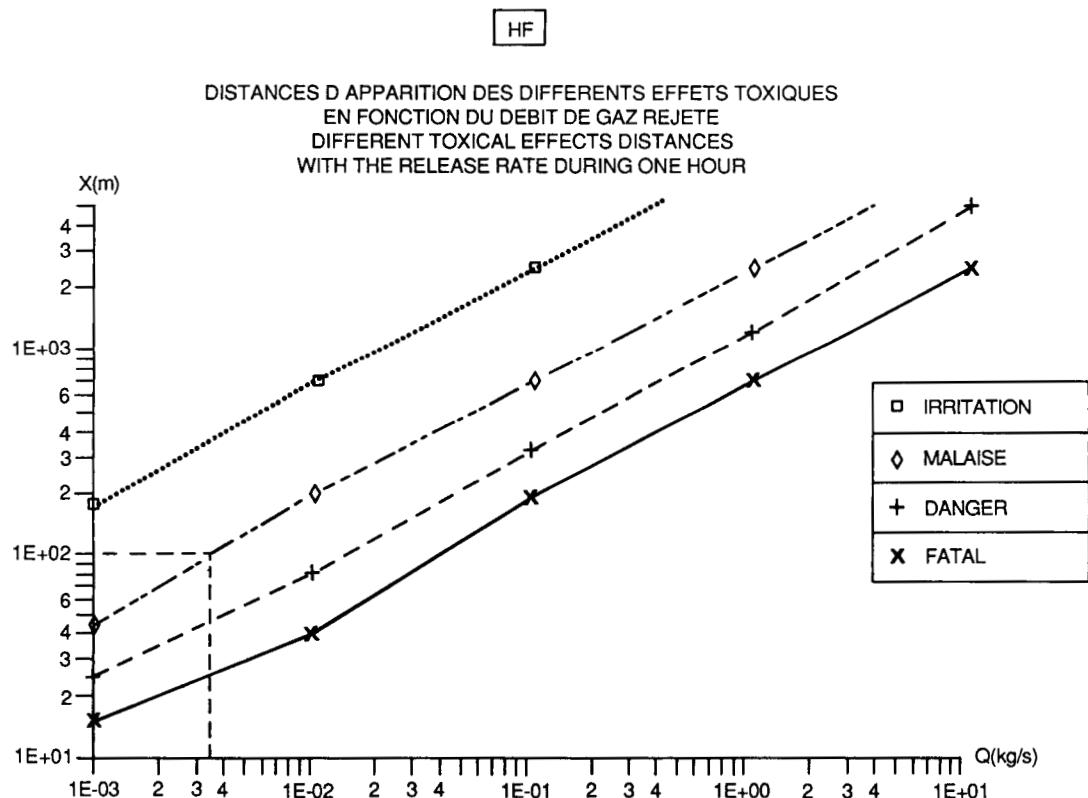
*ALL THE QUANTITY OF FLUOR IS TAKEN IN ACCOUNT (UO₂F₂ AND HF)

Figure 3



Reject pendant 30 mn au niveau du sol.
Vitesse du vent : 5m/s .Diffusion : Normale .
RELEASE DURING THIRTY MINUTES AT THE GROUND LEVEL
WIND SPEED 5m/s – NORMAL DIFFUSION CONDITIONS

Figure 4



Reject pendant 1 h au niveau du sol.
Vitesse du vent : 5m/s .Diffusion : Normale .
RELEASE DURING ONE HOUR AT GROUND LEVEL
WIND SPEED 5m/s - NORMAL DIFFUSION CONDITIONS

Figure 5

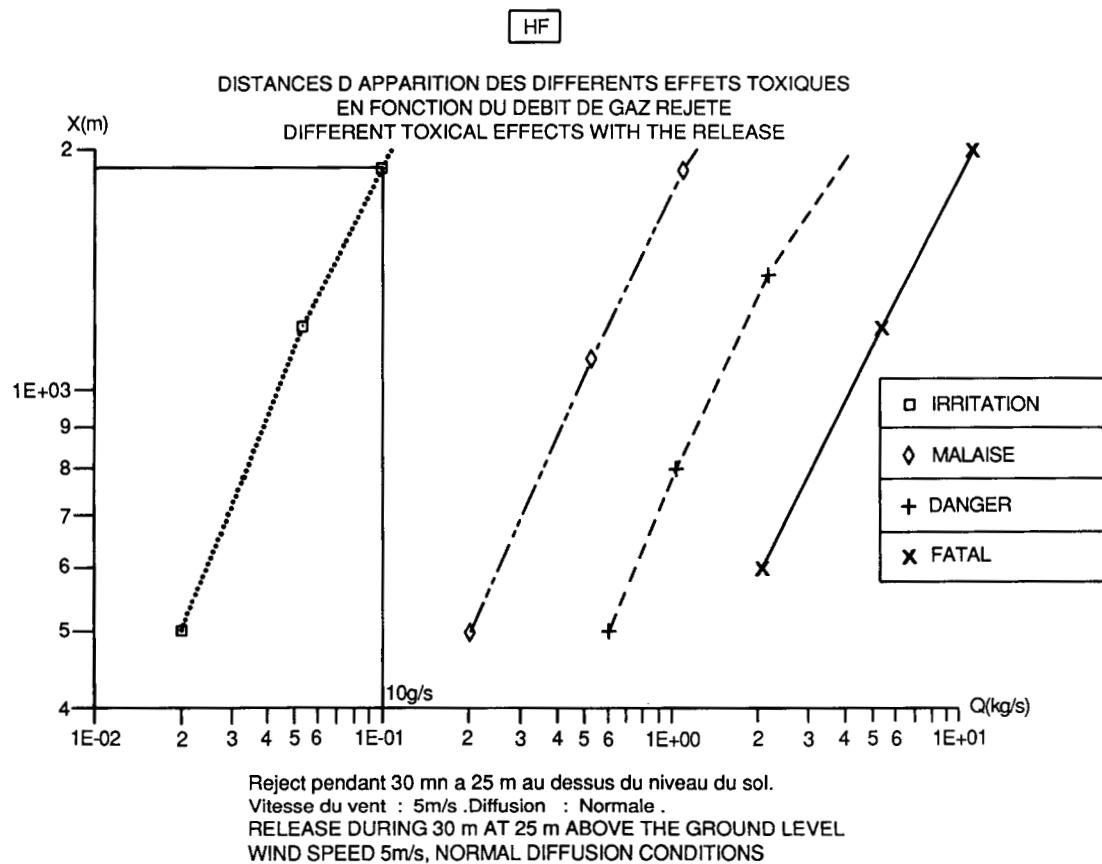


Figure 6

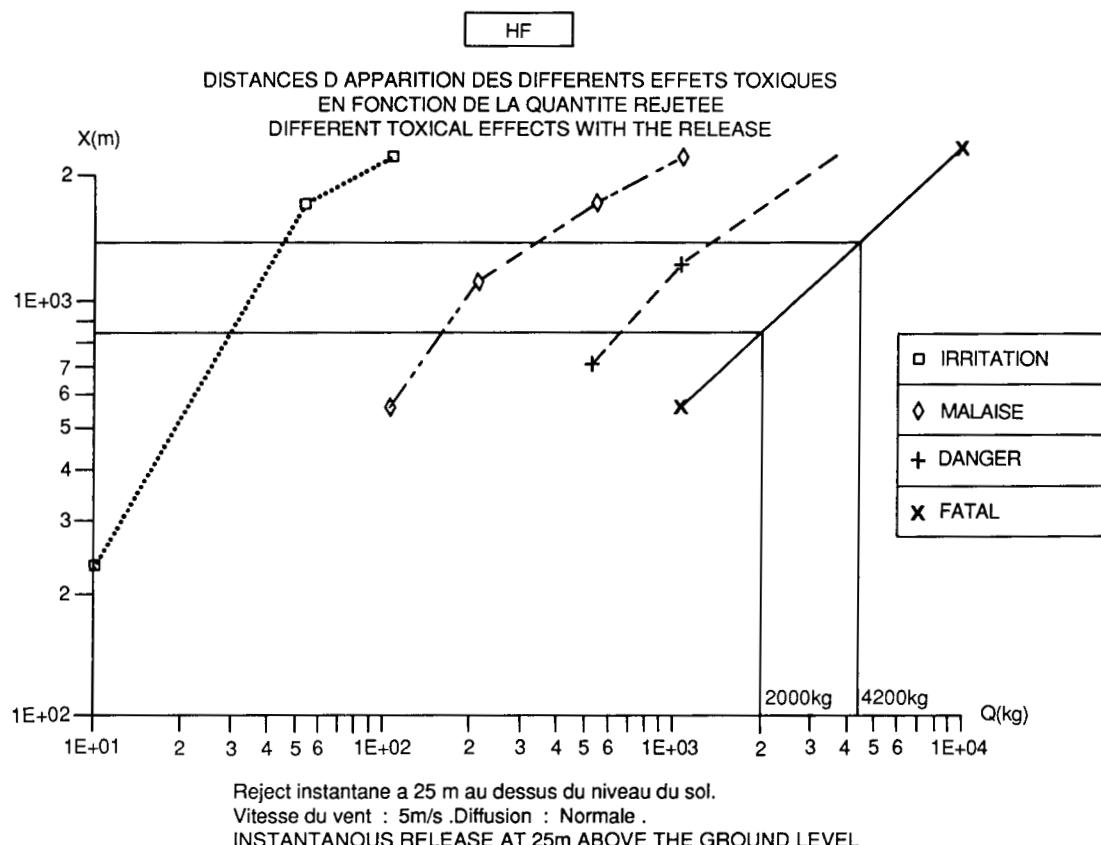


Figure 7

SAFETY-RELATED EVENTS AT U.S. GASEOUS DIFFUSION PLANTS

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ABSTRACT

This report contains material presented in the lecture entitled "Safety-Related Events at U. S. Gaseous Diffusion Plants" as part of the Uranium Hexafluoride-Safe Handling, Processing, and Transporting Conference held in Oak Ridge, Tennessee, May 24-26, 1988. Portions of the material have been previously presented in a paper of the same title as part of the Department of Energy/Argonne National Laboratory Training Course on Prevention of Significant Nuclear Events.

INTRODUCTION

There are three gaseous diffusion plants in the United States, located at Oak Ridge, Tennessee; Paducah, Kentucky; and Portsmouth, Ohio (See Figure 1). The primary function of these plants is to enrich uranium hexafluoride in the uranium-235 isotope for both commercial power reactor and military uses. The plants are

government-owned/contractor-operated with Martin Marietta Energy Systems, Inc., as prime contractor to the United States Department of Energy for all three plants.

The Oak Ridge plant was shut down and placed in ready standby in August 1985 due to the continuing decline in the demand for enriched uranium. The plant was placed in a shutdown mode in 1987. The Paducah and Portsmouth plants are interrelated in the current operating scheme in that slightly enriched product from Paducah is used as one of the feed materials for the Portsmouth plant.

An outline of this report is shown in Table 1. First, a brief description of the gaseous diffusion process is presented, and then two significant, non-critical operating incidents are discussed. Handling of liquid and gaseous uranium hexafluoride (UF_6) is then discussed and two significant incidents involving UF_6 handling are described.

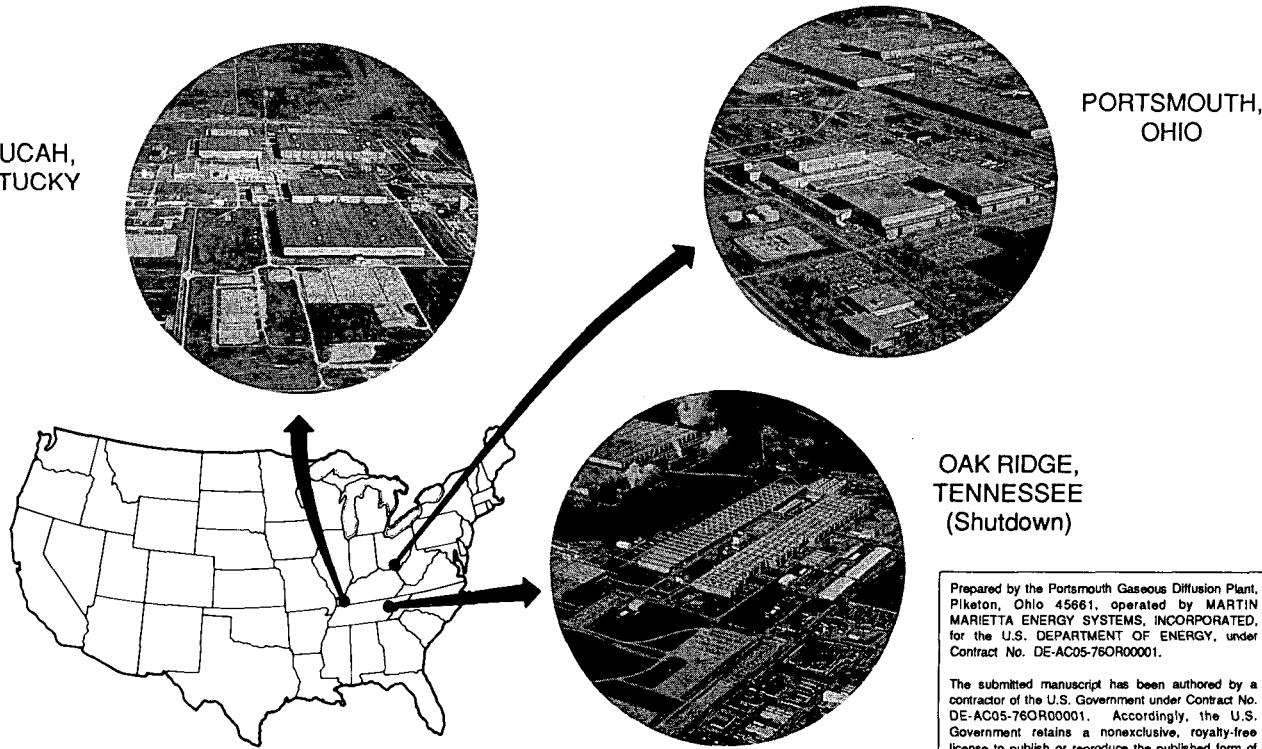


Fig. 1. Gaseous diffusion plants

Table 1. Safety-related events at U.S. Gaseous Diffusion Plants

- PROCESS OVERVIEW
- NUCLEAR CRITICALITY SAFETY
- URANIUM HEXAFLUORIDE HANDLING

PROCESS OVERVIEW

The gaseous diffusion process takes advantage of the slight difference in molecular weights of $U^{235}F_6$ and $U^{238}F_6$ in order to accomplish the isotope separation. As shown in Figure 2, a feed stream of UF_6 is pumped into a diffuser, a large tank-like device containing thousands of porous barrier tubes. Process parameters are maintained to force almost exactly one-half of the feed stream through the walls of the porous barrier tubes while the other half passes down through the tubes and exits through the tube ends. Since the $U^{235}F_6$ molecule is slightly lower in molecular weight than the $U^{238}F_6$ molecule, the normal molecular motion of the lighter molecule within the gaseous mixture is more rapid. Since the lighter molecule is moving faster it is more likely to hit the walls of the porous tubes and, thus, more likely to pass through the walls of the tubes. As a result, the half of the feed stream which passes through the walls of the tubes is slightly enriched in the U^{235} isotope. The half of the feed stream which passes down through the length of the tubes is slightly depleted in the U^{235} isotope.

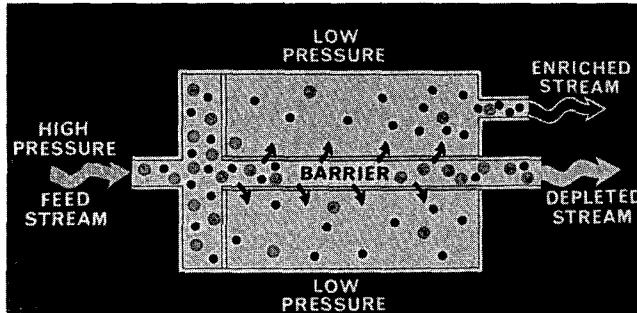


Fig. 2. Gaseous diffusion stage

Since the amount of isotope separation accomplished by a single gaseous diffusion stage is quite small, many stages must be operated in series or "cascaded" in order to effect the desired separation. Cascading is shown schematically in Figure 3. Although the assay numbers shown in Figure 3 are not technically correct, they show the principle of cascading. A stage, in addition to a diffuser, contains a compressor to pump the gas, a motor to drive the compressor, a process cooler, process control valve, and interconnecting piping.

The enriched stream from a stage enters a compressor which pumps the gas to the next upstream stage where further enrichment is performed. Sufficient stages are operated in series to produce the desired product. The depleted stream from a stage enters a compressor which pumps the gas to the next downstream stage where further separation occurs. Sufficient stages are operated below the feed point to produce the desired tails assay. The desired product assay is determined by customer orders while the desired tails assay is determined by economic considerations.

Stages are further grouped into cells, the smallest number of stages which can be taken offstream and shutdown for operational or maintenance purposes. The configuration shown in Figure 4 shows ten 8-stage cells which are typical of the largest equipment in the gaseous diffusion facilities. Cells contain large block valves which permit isolation of the cell equipment from the remainder of the process, and units are simply groups of cells which share certain auxiliary features.

Uranium hexafluoride is processed in the gaseous state throughout the separation process and, in most instances, is below atmospheric pressure. Ton quantities of higher pressure UF_6 are handled, however, in the feed and withdrawal areas of the process.

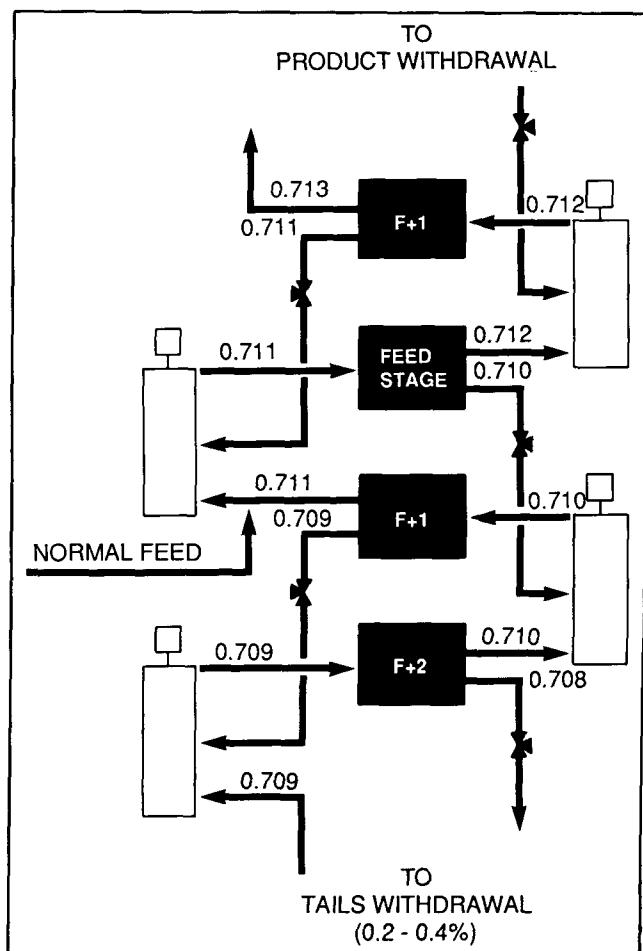


Fig. 3. Gaseous diffusion stage "cascading"

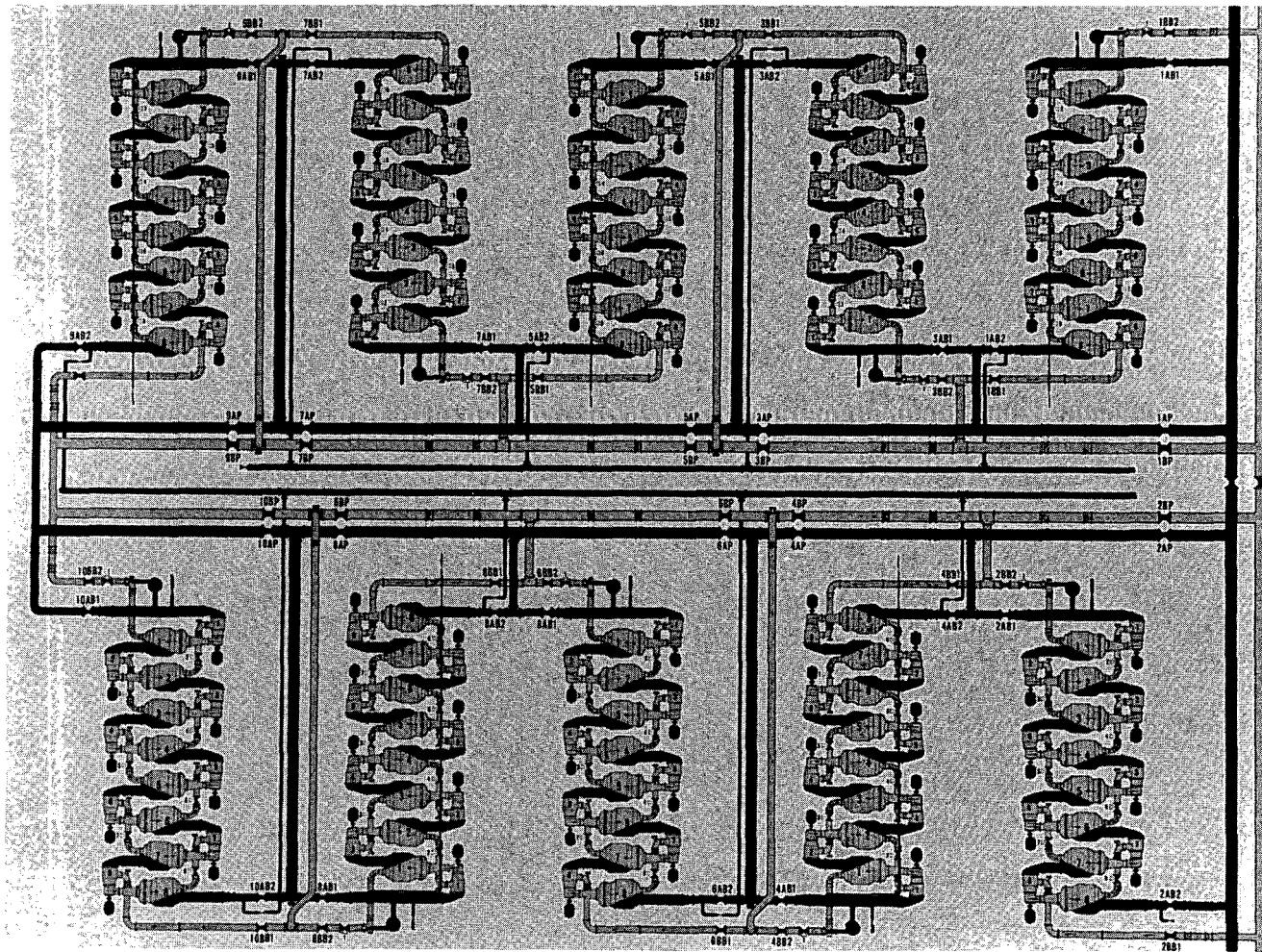


Fig. 4. '000' Unit configuration

NUCLEAR CRITICALITY SAFETY

As seen in the outline in Table 2, there has never been a nuclear criticality accident at the U.S. Gaseous Diffusion Plants. The gaseous diffusion equipment is sized such that the process is critically safe for gases, but may not be critically safe for solid materials. Where uranium must be handled in the solid or liquid form, moderation is controlled and equipment is designed for the processing of the more dense material. We operate at all times under double contingency rules, ensuring that at least two barriers must be breached in order for a criticality to occur.

Although a criticality has never occurred at the diffusion plants, there have been instances where significant quantities of enriched materials have accumulated in the process under uncontrolled conditions. Two of these incidents are described in more detail.

Both enhanced employee training and increased numbers of audits and reviews by the criticality safety groups have resulted from past incidents and operating experiences in the diffusion plants.

Table 2. Nuclear criticality safety

- THERE HAS NEVER BEEN A NUCLEAR CRITICALITY ACCIDENT AT THE U.S. GASEOUS DIFFUSION PLANTS
- GASEOUS DIFFUSION EQUIPMENT IS SIZED SUCH THAT IT IS CRITICALLY SAFE FOR GASES BUT MAY NOT NECESSARILY BE SAFE FOR SOLID COMPOUNDS
- DOUBLE CONTINGENCY RULES
- EMPLOYEE TRAINING
- AUDITS AND REVIEWS BY CRITICALITY SAFETY PERSONNEL

Significant Past Non-Critical Events

Two significant non-critical events are discussed in this report. The first occurred at the Portsmouth Gaseous Diffusion Plant in 1973 and is outlined in Table 3. The incident occurred in the high assay portion of the plant (approximately 97 percent U235) during routine operations. The process control valves in this portion of the process are hydraulically operated using hydrocarbon oil.

In this incident, a 1/4-inch diameter copper process pressure line was physically contacting a similar 1/4-inch diameter hydraulic oil line to a stage control valve. As a result of normal process vibrations, mutual abrasion resulted in small holes in both lines. With holes in both lines, oil from the hydraulic system entered the pressure sensing line and travelled to the below-atmospheric process system. Once inside the process system, the oil reacted with UF6 forming solid compounds. The slow leakage to process continued for an undetermined period of time. In time, the cell began to behave in an unusual manner, and process control became virtually

impossible. Abnormal control valve behavior coupled with high gamma radiation readings from the cell indicated a solid uranium deposit in the piping and one compressor. Neutron probe measurements yielded an estimate of a sizable deposit.

The cell was removed from service, and routine efforts to remove the deposit by in-place chemical treatment were unsuccessful. The piping from which the high gamma was emanating was removed, and a large, moderated deposit was discovered in the 8-inch diameter pipe and accompanying compressor. Photographs of the deposit inside the pipe are shown in Figures 5 and 6.

The area was roped off, and following planning sessions, cadmium strips were inserted, and the material was removed and placed in always-safe containers. Approximately 25 pounds of U235 were recovered.

Recommendations included the inspection and separation of the thousands of instrument and hydraulic lines throughout the process, stressing the importance of using radiation readings to detect uranium deposits, and additional training of supervision and operators.

The second event occurred at the Oak Ridge Gaseous Diffusion Plant in 1981 and is summarized in Table 3. As a result of an error on a drawing, a four-inch line to process was cut allowing a large inrush of atmospheric air to enter the process. This resulted in surging of the axial-flow compressors and failure of one of the compressors. Overheating of the compressor caused growth of the aluminum rotor relative to the steel shell, and metal-to-metal rubbing occurred. Friction resulted in localized elevated temperatures initiating a UF6-metal reaction. The reaction continued and eventually spread to the cooling system which released refrigerant-114 to process, further contributing to the chemical reaction; and the resulting high pressure ruptured an expansion bellows in the cell, relieving the gases to atmosphere.

The UF6-metal reaction reduced the UF6 to a solid uranium compound, and a significant amount of solid material was contained within the cell after the reaction was complete. The U235 assay was approximately 3 percent. Sprinkler systems near the affected cell were removed from service to assure that no water was added to the reaction products. A specially-modified emergency fire truck containing boronated water was available but was not needed. A detailed action plan was developed for removal of the solid deposit. Gamma measurements indicated no real problem, and the equipment was removed, and the deposit was placed into always-safe containers. Approximately 250 pounds of uranium were recovered from the solid deposits from the cell.

Recommendations included inspection and correction of compressor running clearances to allow for rotor growth under unusual circumstances, additional instrumentation installation, review of other similar facilities, and additional training. The additional training included emergency drills involving high temperature reactions within process equipment.

Table 3. Significant past non-critical events

<u>1973</u>
<ul style="list-style-type: none">• PRESSURE SENSING LINE FROM CONTROL VALVE RUBBED AGAINST HYDRAULIC OIL LINE. MUTUAL ABRASION RESULTED IN SMALL HOLES IN BOTH LINES.• OIL ENTERED THE PRESSURE-SENSING LINE AND REACTED WITH UF6.• LARGE, MODERATED DEPOSIT DEVELOPED IN 8-INCH DIAMETER PIPE. ENRICHMENT WAS GREATER THAN 97 PERCENT.• UPON REMOVAL, THE LINE WAS FOUND TO CONTAIN A BLACK, TARRY SUBSTANCE. CADMIUM STRIPS WERE INSERTED.• THE URANIUM-CONTAINING SUBSTANCE WAS REMOVED AND PLACED IN ALWAYS-SAFE CONTAINERS.
<u>RECOMMENDATIONS (1973)</u>
<ul style="list-style-type: none">• INSPECT AND SEPARATE ALL INSTRUMENT AND HYDRAULIC LINES.• IMPORTANCE OF USING RADIATION READINGS TO DETECT AND LOCALIZE URANIUM DEPOSITS WAS STRESSED.• TRAINING.
<u>1981</u>
<ul style="list-style-type: none">• LARGE INRUSH OF ATMOSPHERIC AIR ENTERED PROCESS.• RESULTING COMPRESSOR SURGING AND FAILURE CAUSED COMPRESSOR ROTOR GROWTH.• METAL-TO-METAL RUBBING OCCURRED AS A RESULT OF ROTOR GROWTH.• FRICTION RESULTED IN LOCALIZED ELEVATED TEMPERATURES INITIATING UF6-METAL REACTION.• REACTION SPREAD TO COOLING SYSTEM R-114 WHICH FURTHER CONTRIBUTED TO CHEMICAL REACTION.• HIGH-CELL PRESSURE RESULTED IN PIPING BELLOWS FAILURE.• APPROXIMATELY 250 POUNDS OF URANIUM WAS DISCOVERED FROM SOLID DEPOSITS INSIDE THE CELL.
<u>RECOMMENDATIONS (1981)</u>
<ul style="list-style-type: none">• INSPECT AND CORRECT COMPRESSOR CLEARANCES TO ALLOW FOR ROTOR GROWTH.• ADDITIONAL INSTRUMENTATION TO MONITOR STAGE TEMPERATURES, VIBRATION LEVELS, MOTOR LOADS, AND PROCESS PRESSURES IS NEEDED.• REVIEW OF OTHER OPERATING FACILITIES.• TRAINING.

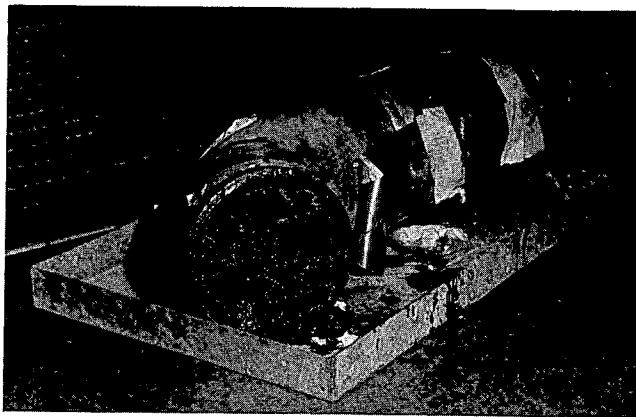


Fig. 5. Process pipe containing UF₆/hydrocarbon reaction products



Fig. 6. Process pipe containing UF₆/hydrocarbon reaction products

Employee Training

Salaried and hourly employees who handle fissile materials receive classroom training annually on the general principles of criticality safety. Both the employee and his supervisor then sign the form shown in Figure 7 following training. Specific on-the-job training in criticality safety is accomplished in the field in addition to the classroom work.

Audits and Reviews by Criticality Safety Professional Staff

Figure 8 shows the form utilized by criticality safety personnel in performing routine, in-plant audits. The audit schedule is computerized and outlines the monthly audit requirements. Following the audit, the incident report is completed and presented to plant supervision responsible for the area audited. Findings with severity indices of 2 and above are also presented to plant management. Audit severity indices are statistically studied to determine trends and areas needing particular attention.

NUCLEAR CRITICALITY SAFETY EMPLOYEE TRAINING REPORT			
NAME	EMPLOYEE NO.	JOB	AREA
DOES HE KNOW AND UNDERSTAND THE FOLLOWING: AS WELL AS OTHER QUESTIONS PERTINENT TO HIS JOB?			
YES NO			
1. Complete operating procedures, assembly sequences, etc.			
2. Emergency signals and evacuation routes.			
3. Criticality emergency procedures.			
4. Restricted use of water in enriched uranium process areas.			
5. Restricted use or complete elimination of genetically unsafe containers in solution areas.			
6. Place and keep lids on containers.			
7. Avoidance of uranium assay crossover; how to identify different assays and work areas.			
8. Modular enriched uranium and non fissile assembly procedures.			
9. Do not place miscellaneous material on, around, or among enriched uranium.			
10. Approved storage positions; how to recognize the proper separation of units (batches) and the limit number of units.			
11. Limits and restrictions of "conditions of approval" for operations.			
12. "Empty" fissile material container handling and storage.			
There must be no deviations in operating procedures; all changes in operating procedures must be formally approved by the Nuclear Criticality Safety Group.			
If there is ever any question or doubt about his job, if he does not know the exact procedure.....			
14. ALWAYS STOP and ask his supervisor.			
REMARKS: (List questions and discussions pertinent to job not covered in above suggestions.)			
The above employee has been properly instructed in nuclear safety rules applicable to his job.		I acknowledge that my supervisor has discussed the above with me, and I understand the nuclear safety requirements of my job.	
SUPERVISOR	DATE	EMPLOYEE	DATE
To be completed immediately for each enriched uranium employee when he enters a new area of responsibility and at least annually thereafter.			

Fig. 7. Nuclear criticality safety employee training report

CRITICALITY SAFETY AUDIT/INCIDENT REPORT																																															
1. DATE OF AUDIT	2. DATE OF DOCUMENTATION																																														
3. INCIDENT FOUND? <input type="checkbox"/> YES (If "YES", Complete Sections 7-18) <input type="checkbox"/> NO (If "NO", Omit Sections 7-18. But Check "D" in Section 18)																																															
4. LOCATION	5. OPERATION <input type="checkbox"/> NONROUTINE <input type="checkbox"/> ROUTINE <input type="checkbox"/> OTHER - SPECIFY																																														
6. NAME OF OPERATION CONTACT																																															
7. DATE OF INCIDENT	8. DETECTION BY: <input type="checkbox"/> PLANT PERSONNEL <input type="checkbox"/> ENGINEER <input type="checkbox"/> OTHER - SPECIFY																																														
9. WHEN INCIDENT DISCOVERED <input type="checkbox"/> AUDIT <input type="checkbox"/> OTHER																																															
10. DESCRIPTION OF INCIDENT																																															
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Fig. 8. Criticality safety incident report

URANIUM HEXAFLUORIDE HANDLING

UF₆ Chemistry

Uranium Hexafluoride (UF₆) is the compound most suitable for use in the gaseous diffusion process. It is a solid at room temperature; however, it can be maintained in the gas phase under controlled temperature and pressure conditions. The phase diagram for UF₆ is presented in Figure 9. The phase relationship is shown as a function of pressure, expressed in pounds-per-square-inch absolute (psia), versus Fahrenheit temperature. The triple point pressure of 22 psia and temperature of 147°F is the only combination of pressure and temperature where all three phases exist simultaneously.

Although UF₆ is a stable compound, it has some chemical properties which add to the handling complexities. It reacts chemically with water to form UO₂F₂ and HF which are water soluble reaction products. The reaction formula is shown at the top of Figure 9. UF₆ is not compatible with organics, such as lubricating oils, and the reaction with hydrocarbon oil can be violent at elevated temperatures. UF₆, which is very corrosive to some metals, is inert to clean, dry metal fluoride films on aluminum, copper, monel, nickel, and aluminum bronze. Mild steel cylinders are acceptable for transport and storage. UF₆ does not react with oxygen, nitrogen, or dry air, and its reactivity with most saturated fluorocarbons is very low. Aside from nuclear considerations, UF₆ can be handled safely in essentially the same manner as any other corrosive and/or toxic chemical by employing procedures developed to accommodate the characteristics of the material.

The reaction of UF₆ with the water vapor available in the air is the greatest concern during UF₆ releases. UF₆ and the products of its reaction with moisture represent a potential health problem. This health concern involves both onsite personnel and the general public offsite.

Controlling a UF₆ release requires the use of emergency procedures and equipment. Respiratory protection equipment, wooden plugs, patches, release detection and alarm systems, and cooling mechanisms are available in the areas where UF₆ is handled. Entry into dense clouds produced by a UF₆ release requires breathing apparatus capable of preventing inhalation of hydrogen fluoride and particulates of uranium compounds. Skin protection is also necessary to prevent burns. All persons not properly protected are evacuated immediately from areas affected by such a release.

Another physical characteristic which must be considered when UF₆ is handled in cylinders is the density change when the material changes from solid to liquid and the continuing liquid density change as a function of temperature. This physical characteristic is shown in Figure 10. There is an approximate 30 percent decrease in density (increase in volume) as solid UF₆ melts and a continuing increase in volume as the liquid is heated. Cylinders are normally filled in the diffusion plants at temperatures approximating 160°F, and since several hours are usually required to fill a cylinder, some solidification of the UF₆ can occur during filling. Thus it is possible to fill a cylinder with more UF₆ than it can phys-

ically hold at elevated temperatures. Cylinder fill limits are generally established ensuring a 3-5 percent void volume at 250°F. Exceeding the cylinder fill limit and/or heating the cylinder above 250°F can cause the UF₆ to expand to fill all of the free volume. Further heating will cause the confined liquid to develop hydraulic pressures which can rupture the cylinder. This is a well understood phenomenon, and its occurrence is prevented by strict adherence to cylinder fill limits and temperatures.

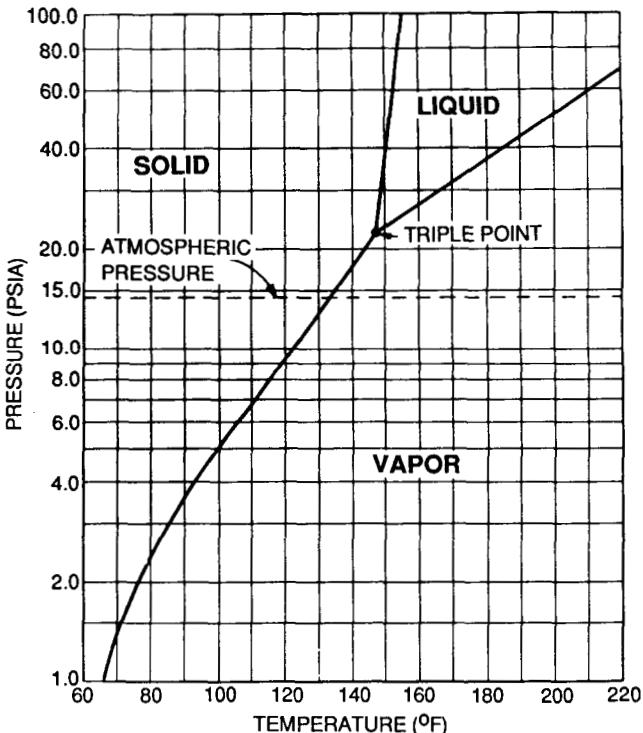


Fig. 9. Phase diagram for uranium hexafluoride

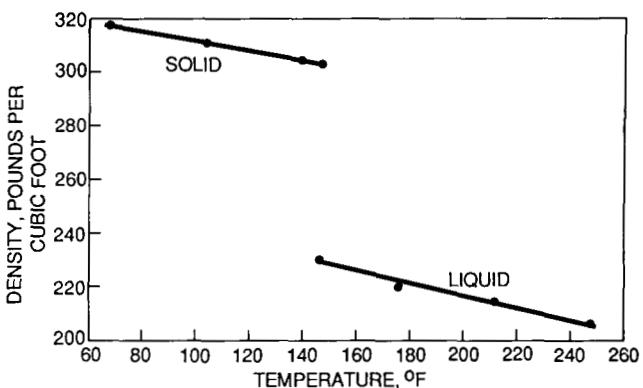


Fig. 10. Densities of solid and liquid UF₆

SAFETY CONCERN

Safety Goals

The potentially toxic effects of UF₆ and the products of the UF₆-water reaction mandate that UF₆ releases be minimized or eliminated. The UF₆ sampling, feeding, and withdrawal operations are performed at elevated temperatures and pressures. In addition, some cylinder moves are performed while the cylinder contains hot, liquid UF₆. Therefore, studies to determine the past causes of UF₆ releases from these operations, to determine other credible release scenarios, and to prevent and/or mitigate UF₆ releases have become high priority activities at the gaseous diffusion plants. In general, these studies did not address criticality, contamination, fire safety, or general industrial safety. These concerns are already addressed by other staff organizations.

General Accident Scenarios

The accident scenarios that could lead to significant UF₆ releases fall into three general categories. These are equipment failure, operator error, and natural phenomena. These are detailed in Table 4.

UF₆ releases have occurred at all three gaseous diffusion plants over a hundred years of combined operation. The accidents that have resulted in UF₆ releases of 5 kilograms or more are listed in Table 5. A summary of causes of the significant UF₆ releases is detailed in Table 6.

Cylinder Heating Safety Systems

Gaseous diffusion plant management recognized early that if a very serious UF₆ release were to occur it would occur in the areas where large quantities of UF₆ are handled in liquid form, and in particular, those areas where UF₆ is heated, liquified, sampled, and vaporized would be the most likely to suffer a severe problem. As a result of this recognition, steam-heated autoclaves which are utilized to heat UF₆ cylinders have been equipped with extensive redundant instrumentation.

Figure 11 is a schematic of a fully instrumented autoclave, and Table 7 outlines the safety systems installed in all diffusion plant autoclaves.

Table 4. General accident scenarios

EQUIPMENT FAILURES

- MECHANICAL FAILURE OF CYLINDER
- FAILURE OF CYLINDER HANDLING EQUIPMENT
- CYLINDER VALVE OR PIGTAIL FAILURE
- FAILURE OF MANIFOLD OR VALVES

OPERATOR ERROR

- MISOPERATION OF CYLINDER HANDLING EQUIPMENT
- IMPROPER PROCEDURE DURING SAMPLING, TRANSFER, FEED OR WITHDRAWAL OPERATIONS
- NATURAL PHENOMENA - FLOOD, WIND OR TORNADO, OR EARTHQUAKE

Table 5. Significant UF₆ releases in U.S. Gaseous Diffusion Plants
1960 - Present

LOCATION	DATE	DESCRIPTION
FEED VAPORIZATION	11/60	HYDRAULIC RUPTURE OF CYLINDER - OPERATOR ERROR
TAILS WITHDRAWAL	11/60	PIGTAIL FAILURE
LABORATORY FEED VAPORIZATION	11/61	CYLINDER VALVE FAILURE
FEED VAPORIZATION	7/65	PIGTAIL RUPTURE
TAILS STORAGE	3/66	LIQUID CYLINDER DROPPED AND RUPTURED CYLINDER WALL - OPERATOR ERROR
LIQUID FEED SAMPLING	5/69	PIGTAIL EVACUATION WITH CYLINDER VALVE OPEN - OPERATOR ERROR
TAILS WITHDRAWAL	7/69	CYLINDER VALVE WOULD NOT CLOSE
FEED VAPORIZATION	12/70	PIGTAIL CONNECTION LEAK
FEED VAPORIZATION	12/70	PIGTAIL LEAK
SAMPLING	1/71	BROKEN PIGTAIL - OPERATOR ERROR
TEST LOOP FEED VAPORIZATION	4/71	PIGTAIL CONNECTION FAILED
SAMPLING	5/73	CYLINDER VALVE WOULD NOT CLOSE
LIQUID PRODUCT TRANSFER	9/75	CYLINDER FAILURE RESULTING FROM EXPLOSIVE UF ₆ OIL REACTION
SAMPLING	11/75	PIGTAIL CONNECTION LEAK
TRANSFER BAY	9/76	PIGTAIL CONNECTION LEAK
TRANSFER AUTOCLAVE	12/77	CYLINDER VALVE THREAD LEAK
FEED VAPORIZATION	12/77	PIGTAIL CONNECTION LEAK
TAILS WITHDRAWAL	1/78	RUBBING COMPRESSOR - UF ₆ /R-114 REACTION
SAMPLING	3/78	CYLINDER CONNECTION LEAK - OPERATOR ERROR
LIQUID CYLINDER STORAGE	3/78	CYLINDER RUPTURE FROM STRADDLE CARRIER DROP
TAILS WITHDRAWAL	10/78	CYLINDER VALVE BROKE AS RESULT OF TRANSPORT WHILE CONNECTED - OPERATOR ERROR
X-326	7/82	TWO PURGE CELLS SHUT DOWN - INSUFFICIENT EQUIPMENT OPERATING TO SEPARATE UF ₆ FROM AIR
X-326	12/83	REVERSE PRESSURE DIFFERENTIAL IN PURGE CASCADE RECYCLE
X-333	1/86	OVERLOADED TRAPS - FAULTY MONITORING DEVICE ALLOWED TRAPPING SYSTEM TO OVERLOAD AND VENT TO ATMOSPHERE

Table 6. Summary of failures

FAILURE TYPE	NUMBER
PIGTAIL	8
OPERATOR ERROR	8
CYLINDER VALVE	4
CYLINDER DROP AND RUPTURE	1
CYLINDER RUPTURE UF ₆ -OIL	1
EQUIPMENT FAILURE	2

SIGNIFICANT SAFETY-RELATED UF₆ HANDLING EVENTS

Two of the more significant accidents, one involving a liquid UF₆ product cylinder and the other involving a liquid UF₆ feed cylinder, demonstrate the need for UF₆ release prevention and/or mitigation.

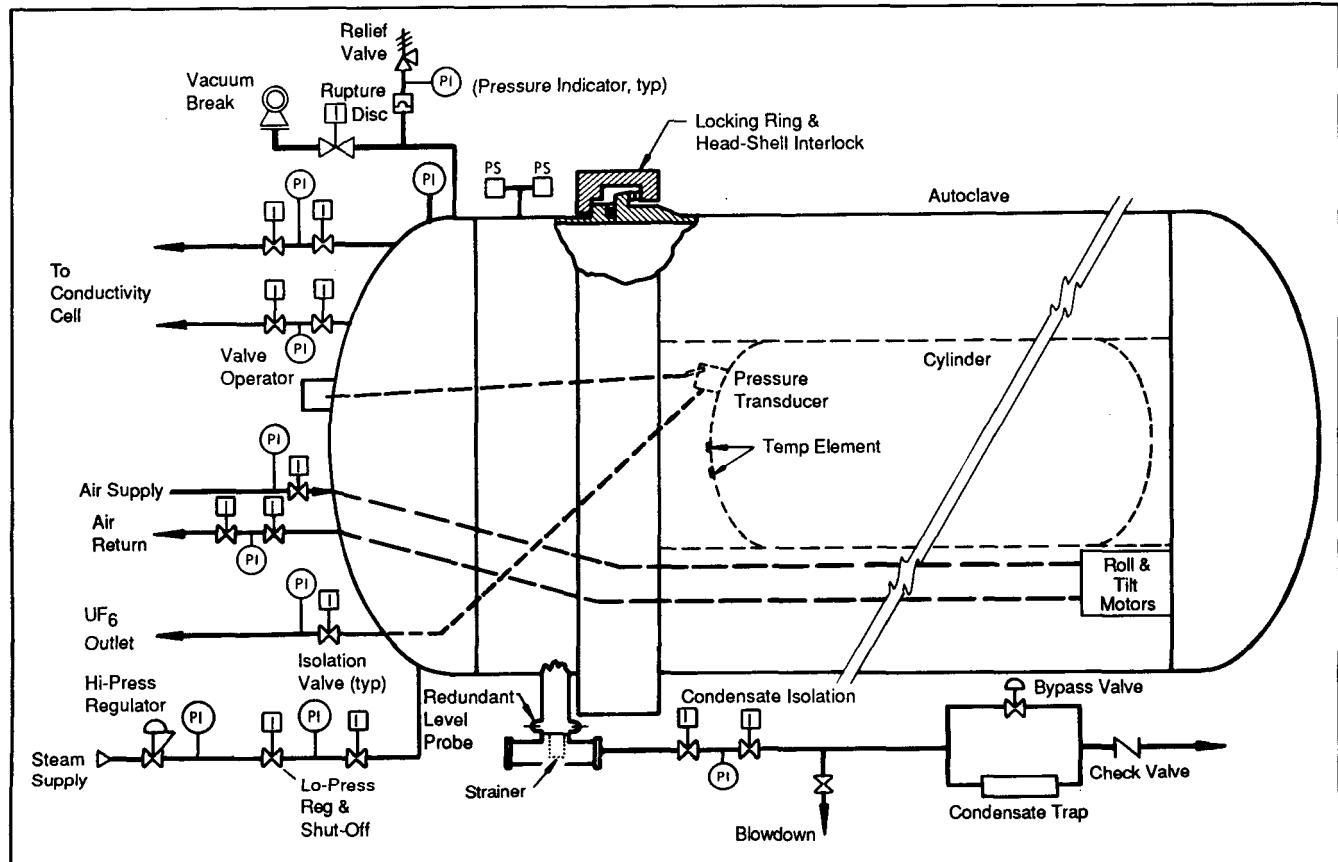


Fig. 11. Autoclave schematic

Table 7. Sampling autoclave safety system

SAFETY SYSTEM	ACCIDENT CONDITION	DESCRIPTION
CRITICALITY CONTROL	NUCLEAR CRITICALITY	REDUNDANT LIQUID LEVEL PROBES, CLOSES STEAM SUPPLY VALVES
RADIATION MONITORING	NUCLEAR CRITICALITY	VISUAL AND AUDIBLE ALARM LOCALLY AND REMOTELY
AUTOCLOVE PRESSURE CONTROL	AUTOCLOVE RUPTURE	LIQUID LEVEL PROBES, PRESSURE RELIEF VALVE, RUPERTURE DISC
UF ₆ CYLINDER PRESSURE CONTROL	CYLINDER RUPTURE	PRESSURE TRANSMITTER, THERMOCOUPLE, CLOSE STEAM SUPPLY VALVES
UF ₆ RELEASE CONTROL	TOXIC MATERIAL RELEASE	HIGH PRESSURE DETECTORS (CLOSE ISOLATION VALVES), LOW PRESSURE DETECTORS (PREVENT HYDRAULIC SYSTEM FROM BEING ENERGIZED), VALVE CLOSING DEVICES
UF ₆ RELEASE DETECTION	TOXIC MATERIAL RELEASE	OUTSIDE AUTOCLAVE, ISOLATES AUTOCLAVE

Cylinder Rupture, UF₆-Oil Reaction

The accident involving the UF₆ product cylinder occurred in 1975 at Oak Ridge and was the result of a UF₆-hydrocarbon oil reaction. Oil was introduced into a Model 30A 2 1/2-ton cylinder by a faulty vacuum pump. When the hot, liquid UF₆ was

poured into the cylinder, the resultant high pressure from the reaction caused the concave cylinder heads to bulge and crack. Figure 12 shows one head of the cylinder. The name plate was thrown off with considerable force, and a small amount of UF₆ was released. It is estimated that less than two liters of oil were contained in the cylinder. The estimated amount of energy released by the reaction was 233 kcal (720,000 ft-lb), resulting in an internal cylinder pressure in excess of 1200 psi.



Fig. 12. Damaged 2 1/2-ton cylinder

Recommendations from the incident included the exclusion of oil filled vacuum pumps for evacuation of UF₆ cylinders. The evacuation of cylinders must be accomplished with equipment such as air ejectors. Also, quality assurance plans were prepared for cylinder decontamination, evacuation, and valve installation.

Findings and recommendations from this incident were made available to nuclear industry licensees and vendors involved in UF₆ handling. This incident is documented in report K/P-6197, Revision 1, "Investigation of a Uranium Hexafluoride Release Incident on September 17, 1975, in the K-1423 Toll Enrichment Facility," April 16, 1986.

Fourteen-Ton Feed Cylinder Drop

An accident involving a 14-ton feed cylinder occurred in 1978. Details of this release are given in Figures 13 and 14. A 14-ton cylinder containing liquid feed material was being moved from the sampling area at Portsmouth to the cooling yard. As the cylinder was being lowered from the straddle carrier to cylinder "saddles," the cylinder fell 8-10 inches resulting in an 8-inch long rupture in the cylinder wall. Essentially, the entire contents of the cylinder were released.

Recommendations included design and installation of a straddle carrier lift mechanism which

provides positive attachment to cylinders, improved straddle carrier maintenance procedures, and improved training in cylinder handling.

CONCLUSIONS

Conclusions and lessons learned are summarized in Table 8. For many years we have dismissed accidents as "operator error," disciplined the operator, and considered the case closed. Over the years we have learned that most "operator errors" are management inspired by poor procedures, poor examples, and/or lack of proper training courses, both classroom and on-the-job. Recent studies have shown that approximately 85 percent of problems are "system problems" with only 15 percent qualifying as true people problems. Recognition of this fact and tackling the system problems instead of finger pointing at individuals is the first step toward improved facility safety. Focusing on system problems includes training the work force thoroughly, audits by health and safety disciplines, visible and enthusiastic upper management support of health and safety, open communication, both vertically and horizontally, and learning from past experiences. Improving the overall system can have a marked effect on facility safety performance.

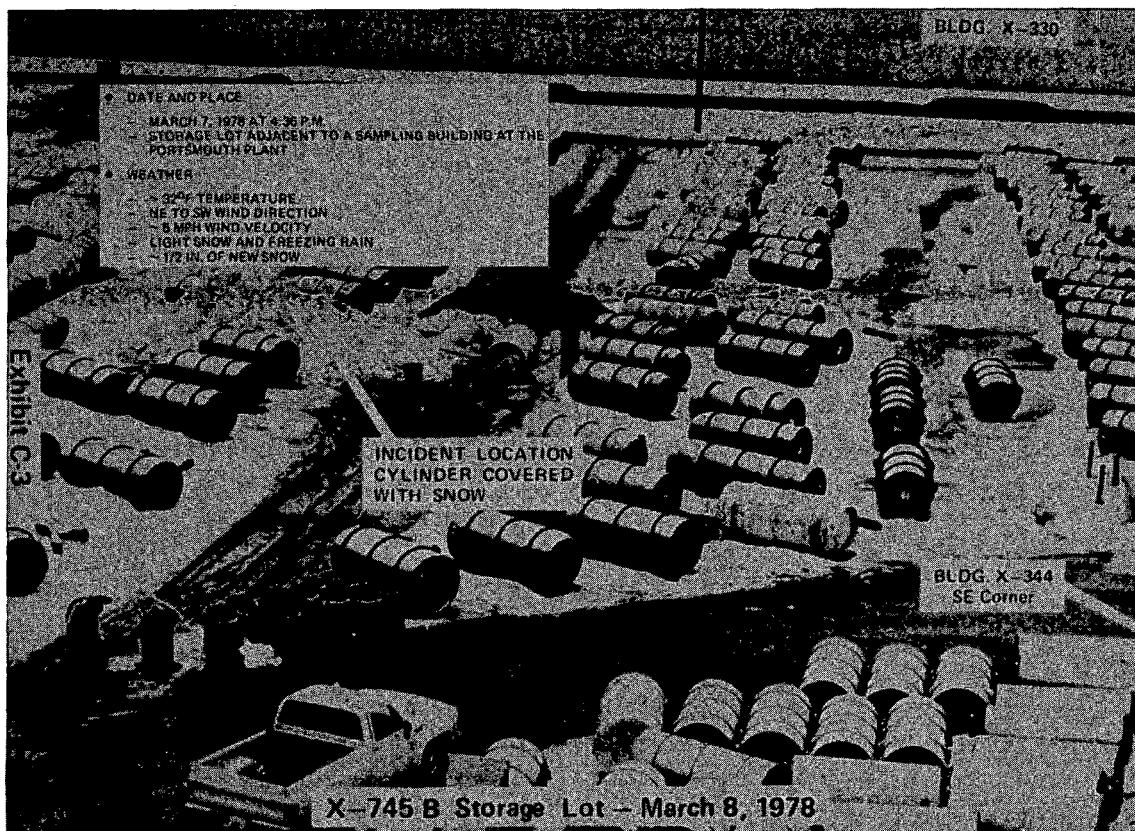


Fig. 13. Large UF₆ release from a 14-ton cylinder

- CIRCUMSTANCES
 - A STRADDLE CARRIER WAS BEING USED
 - A MODEL 48G CYLINDER CONTAINING LIQUID UF₆ WAS DROPPED FROM ~8 - 10 in. HIGH ONTO A WOODEN SADDLE
 - THIN WALL 5/16 in. THICK
 - NORMAL ASSAY UF₆
 - THREE, 1 in. THICK BY 2 1/2 in. WIDE STIFFENING RINGS
 - THE IMPACT RESULTED IN A RUPTURE IN THE CYLINDER WALL ABOUT 1/2 in. WIDE AND 8 in. LONG

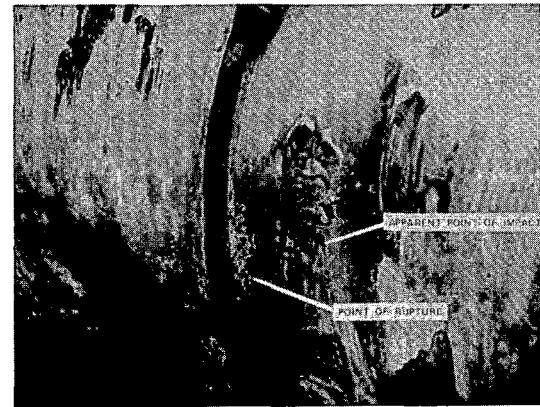
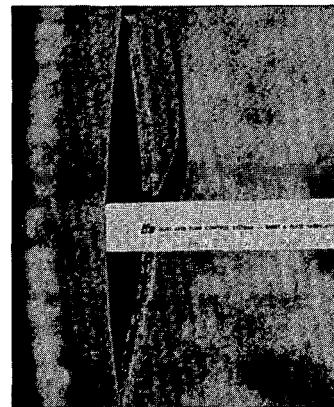
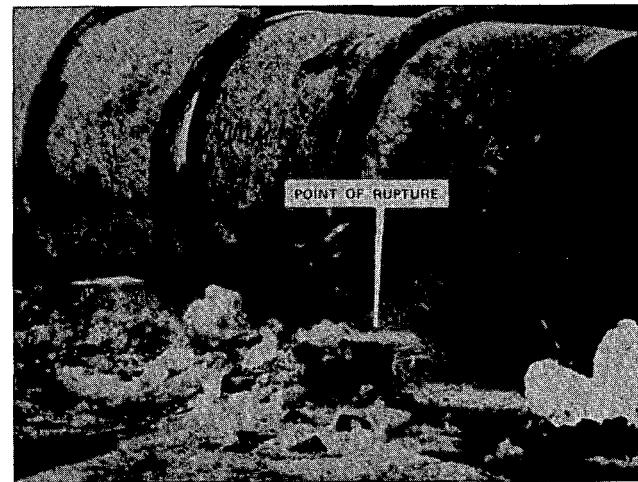


Fig. 14. Point of rupture, large UF₆ release from 14-ton cylinder

Table 8. Conclusions

- 85 PERCENT OF PROBLEMS ARE "SYSTEM PROBLEMS"; 15 PERCENT ARE "OPERATOR ERROR"
- WORKERS AND SUPERVISION MUST BE THOROUGHLY TRAINED IN ALL ASPECTS OF THEIR JOB ASSIGNMENTS
- PLANTWIDE AUDITS BY PRODUCTION PERSONNEL AND HEALTH AND SAFETY DISCIPLINES ARE EXTREMELY IMPORTANT TO ENSURE THAT PROCEDURES ARE ADEQUATE AND ARE BEING FOLLOWED
- MANAGEMENT SUPPORT OF HEALTH AND SAFETY CONCERN MUST BE ENTHUSIASTIC AND CONSPICUOUS
- OPEN COMMUNICATIONS MUST BE MAINTAINED AMONG MANAGEMENT, STAFF GROUPS, AND PRODUCTION EMPLOYEES
- WE MUST LEARN FROM PAST EXPERIENCES

FEEDING UF6 WITHOUT LIQUEFACTION

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ABSTRACT

In a gaseous diffusion enrichment plant, like the GEORGES BESSE plant, uranium hexafluoride has to be transformed from a solid to a gaseous state in order to be fed into the cascade. Two different processes are available.

The first one is based on liquefaction and high pressure evaporation. Its advantages are : firstly sufficient UF6 pressure to directly feed the cascade, and secondly high flow rates. Its one disadvantage comes from the physical properties of UF6 : the liquefaction pressure is overatmospheric (up to 6 absolute bars in some operating conditions), and this creates safety concerns with regards to certain cylinders and other equipment.

The second process simply consists of direct sublimation of UF6. It allows UF6 to stay at subatmospheric pressure, but this low pressure can be incompatible with the cascade feed pressure and the sublimation flow rate is lower than that of evaporation.

This paper describes how a feed process using the low pressure sublimation of solid UF6 has been developed and adapted to a gaseous plant, resulting in net flow rate performances almost equivalent to the ones obtained from a process using UF6 liquefaction.

The solid feed material is shipped using different types of model 48 cylinders. These cylinders are placed in air-heated ovens. The UF6 is sublimated and fed into the cascade through vacuum spiral-pumps.

Two feeding modes by sublimation are being used : in the first one, which is the most used, the oven temperature is 115°C and the net flow rate is 50 grams of UF6 per second and per feed cell (versus 60 g/s for the liquefaction mode). In the second one, the air temperature is limited to 85°C in order to avoid liquefaction of UF6 in any circumstances ; then the net flow rate is only 25-30 g/s of UF6 per feed cell.

This process has received the approval of the Safety Authorities.

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- 1 - Introduction
- 2 - Historical development of the Eurodif feed process
- 3 - Equipment description
 - 3.1. Ovens
 - 3.2. Filters
 - 3.3. Process lines
 - 3.4. Pumps
 - 3.5. Building and miscellaneous equipment
- 4 - Process operation
 - 4.1. Working principles
 - 4.2. Computerized operating procedures
 - 4.3. Monitoring
- 5 - Operating experience
 - 5.1. Process performances
 - 5.2. Equipment
 - 5.3. Lessons learnt from incidents
- 6 - Conclusions.

1- INTRODUCTION

The GEORGES BESSE Gaseous Diffusion Uranium Enrichment plant located in Tricastin, France, has an enrichment capacity of 10.8 million Separative Work Units per year. Begun in 1974 and completed in 1982, the plant is operated by EURODIF PRODUCTION, a subsidiary of EURODIF S.A. which is composed of the following partners : COGEMA (France), SYNATOM (Belgium), ENUSA (Spain), ENEA (Italy) and AGIP (Italy), SOFIDIF (Franco-Iranian) :

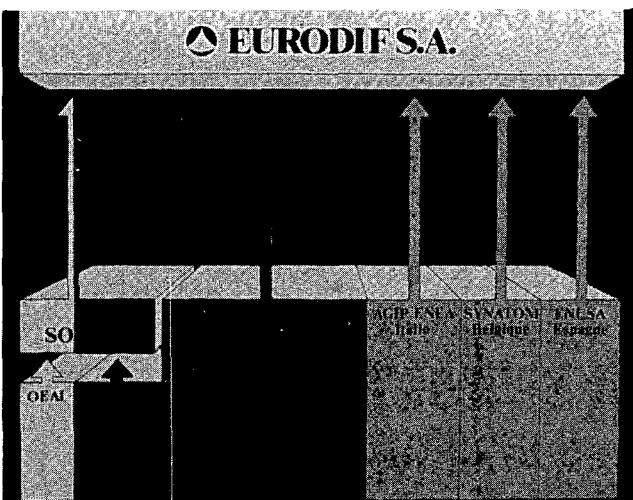


Figure 1. EURODIF S.A.

The cascade feed can reach 35000 metric tons of uranium hexafluoride per year, that is to say 1.1 Kg of UF6 per second. This uranium is mainly natural.

The UF6 cylinders are of different types of model 48 (48 Y, 48 G), coming from various origins around the world. Both the cylinders and their contents remain the property of Eurodif's clients. It can happen that filled cylinders spend long periods in storage yards, before being fed into the cascade. Some of them will have overstayed the hydraulic test deadline. Others, like the 48 G cylinders, have a low steel casing thickness of 8 mm (instead of 16 mm for 48 Y cylinders).

In order to face up to any situation and in spite of the fact that no accident has ever occurred in feed operations, EURODIF decided in 1979 to design an adaptation for its feed process.

2 - HISTORICAL DEVELOPMENT OF THE EURODIF FEED PROCESS

The original installation consisted of 24 air-heated ovens operating at 115°C, liquefying UF6 under a pressure of up to 6 absolute bars. There were several reasons for choosing this particular solution :

- we wanted to avoid using steam heating, because of the risks of criticality, chemical reactions, and external corrosion of cylinders,
- the temperature had to be limited to 115°C in order to avoid decomposition of UF6 when in contact with metals, which happens at temperatures above 120°C,
- this specific number of ovens enabled a total feed rate of 1100 g/s 24 hours a day,
- this type of oven was nearly three times cheaper than autoclaves.*

In 1979, knowing that it was likely that both 48 G and out-of-date cylinders would have to be dealt with, Eurodif designed an adaptation for its feed process. As ovens were already in place, the idea of investing in autoclaves was not selected. It was considered preferable to modify the pressure specifications as follows :

- maximum 1500 absolute mb (triple point of UF6) under any circumstances for 48 G cylinders : mode LPG (low pressure for 48 G).
- maximum 5000 absolute mb (maximum allowed by regulations) for out-of-date 48 Y cylinders : mode LPY (low pressure for 48 Y).

* 210 000 FF compared with nearly 620 000 FF at 1980 values

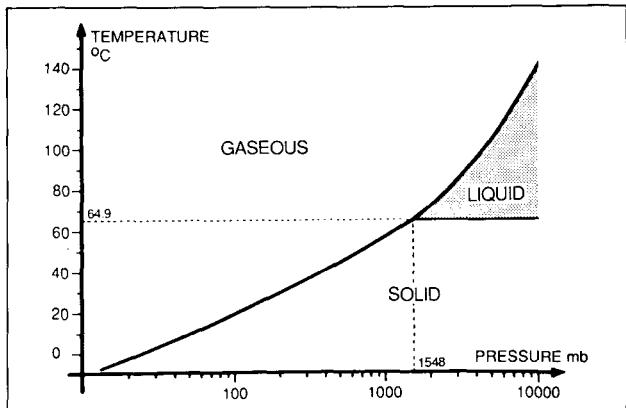


Figure 2. Triple point diagram

Low pressure sublimation was achieved by using a pumping process.

The original feed equipment plus its later improvements were designed and constructed by the same engineering company USSI. The cost of the original unit was about 50 million FF (at 1980 values) without automatic control. Its modification cost 30 million FF, automatic control included (at 1980 values) and was in operation by 1982.

3 - EQUIPMENT DESCRIPTION

3.1. Ovens

Cylinders are heated by means of blown air, using 380 V electric heaters. The amount of heating absorbed by each oven is equivalent of 30 kW. The resulting temperature within the oven can be of up to 120°C.

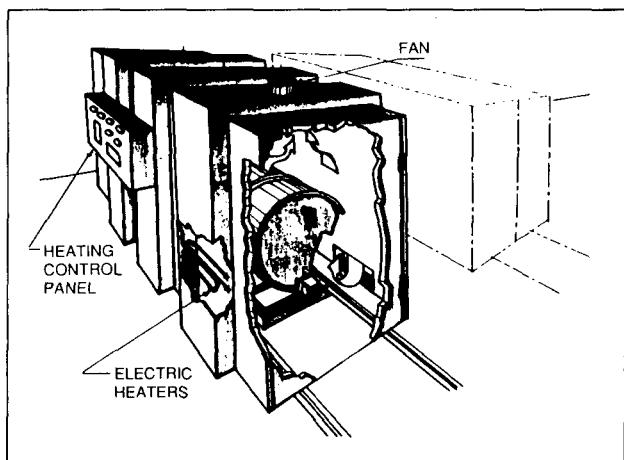


Figure 3. Oven

The oven air temperature is regulated using a controller which has an accuracy of $\pm 1^\circ\text{C}$.

If the temperature exceeds 121°C , a temperature-sensing alarm, which is independent of the control sensor, automatically shuts down the electric heating. An independent pressure sensor reacts in the same way if the pressure goes above 750 mb.

Description : 4.875 meters long, 1.74 meters wide, 2.30 meters high, heat insulation by means of fibre-glass wool, internal walls made of totally welded stainless steel 316.

The cylinder is connected to the feed lines through a mechanical filter.

3.2. Filters

Each filter consists of a nest of tubular filter cartridges inside a cylindrical casing. Each cartridge is made up of 4 standard METAFRAM* tubes welded in line one to the other.

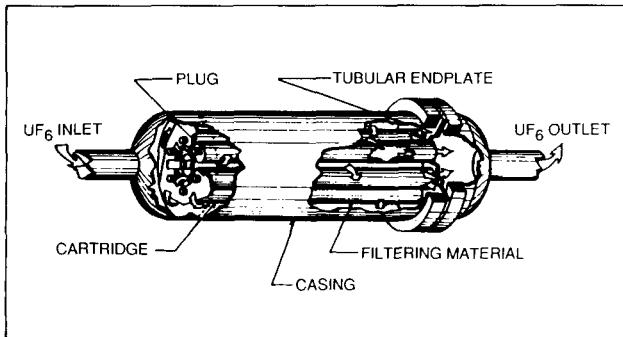


Figure 4. Filter

One end of each cartridge is fixed by expansion into the end plate, the other being sealed.

The UF6 circulates from the exterior towards the interior of the cartridges. Each filter unit can be fitted with an external gamma-ray shield. The cartridge is made of nickel and the casing and flanges are made of stainless steel 304 L. One filter consists of 113 cartridges. The filtration threshold is 2 microns. In feed mode the pressure ranges from 100 to 500 mb.

In evacuation mode, from 100 to 5 mb.

In-filter pressure loss is of 5-6 mb at an operating pressure of 100 mb, a temperature of 115°C and a flow-rate of 79 grams/sec.

3.3. Process lines

3 lines connect the feed unit to the cascade. The two main ones can carry any UF6 flow rate between 20 and 1100 g/s through control valves. The third auxiliary line has a maximum flow rate of only 250 g/s. The pipes are kept warm by means of self regulating electric heating wires (maximum temperature 65°C). The pipes are made of insulated stainless steel.

3.4. Pumps

These are NORMETEX type pumps. 6 pumps are installed. The particularity of these vacuum pumps is that they are completely "dry": UF6 is in contact with neither seal, nor lubricating oil. The tightness is achieved using bellows, which allow the rotation-translation of one spiral inside another, the latter being fixed. Each pump has a flow rate of 115 l/s. The inlet pressure is of a maximum of 200 mb. The discharge pressure is between 550 and 1500 mb. The pump has an automatic cut-off if the discharge pressure reaches 2000 mb (detected by a pressure sensor).

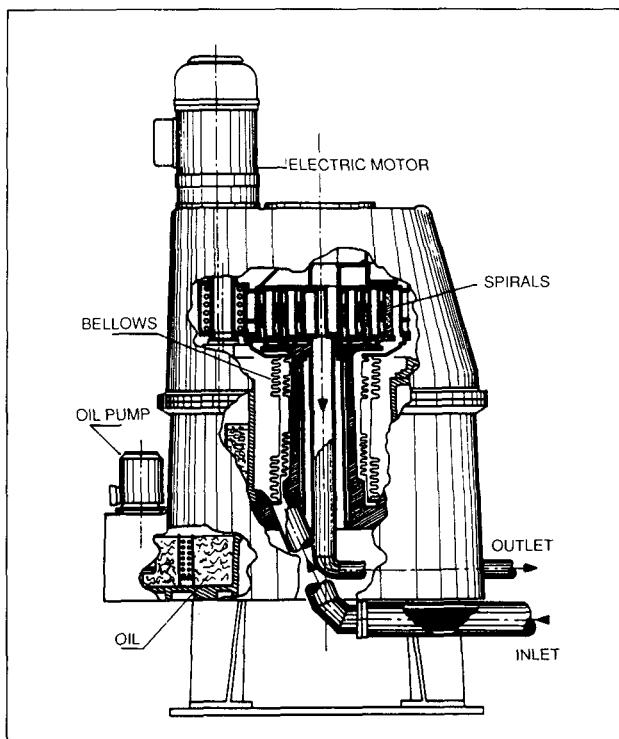


Figure 5. NORMETEX pumps

*METAFRAM : Société de fabrication d'éléments frittés.

3.5. Buildings and miscellaneous equipment

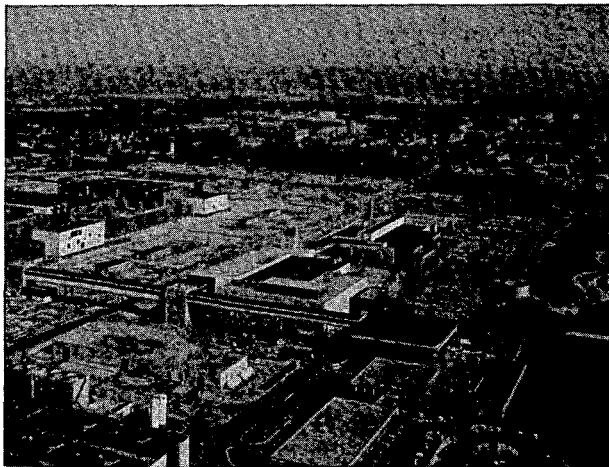


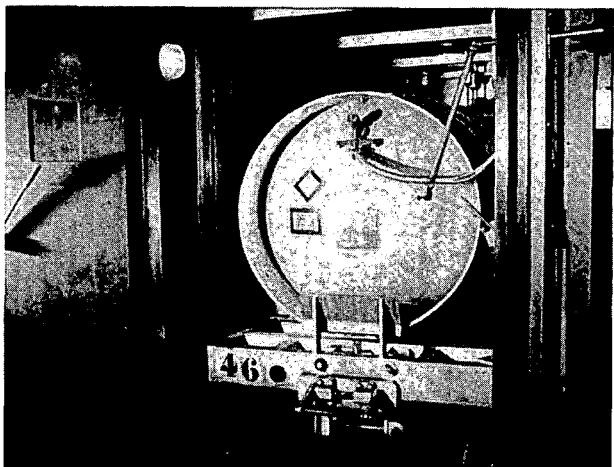
Figure 6. EURODIF site lay-out

The feed unit takes up a part of the building shown on the EURODIF SITE lay out. In this building, feed operations, product and waste withdrawals, cascade purging, vacuum pumping and removal/filling operations are carried out. The building is situated opposite the gaseous diffusion facility to which it is connected by a technical gallery which contains process connection pipes.

The U area contains the different UF6 equipment and circuits, and can be entered only from the intervention zone. An observation and control area is situated between the U - areas and the auxiliary area.

The only pipes containing UF6 which pass through the intervention zone have a pressure of less than atmospheric pressure.

The maximum allowed leak rate for all equipment and piping is 2×10^{-2} lusec * of helium per cubic meter. The feed unit is divided into two zones, that of the ovens and that of the pumps and valves. The 24 ovens are divided into two rows of 12, situated on both sides of a handling area. The pump and valve zone is a separate building of 9 x 22 meters and 9 meters high.



Figures 7 et 8. Feed Unit - The area and an oven

* 1000 LUSECS: One liter per second for a pressure of 1.3 millibar.

4 - PROCESS OPERATION

4.1. Working principles

Each of the 3 feed lines F, G and H can operate either in HP (high pressure) or in LP (low pressure) mode. Operating control is carried out from the central control room which is situated in a different building from that of the feed unit.

The control systems consist of :

- synoptic graphic displays,
- continually-updated on-screen messages
- control software for the ovens and vacuum pumps.

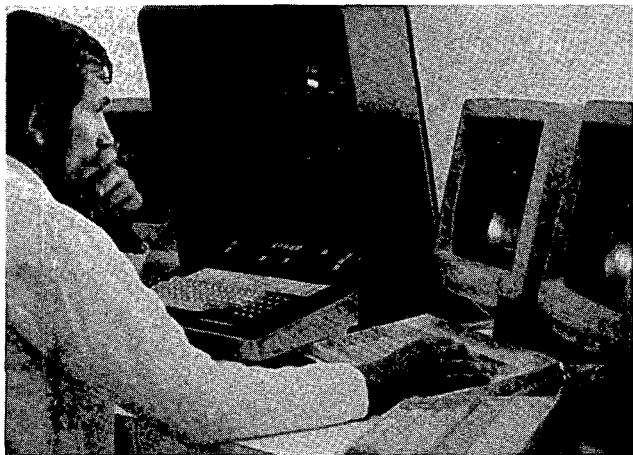


Figure 9. Control room

Connecting and disconnecting of cylinders is carried out on the order of the control room. The necessary dialogue between the system and oven personnel is achieved using a visual indicator and switch panel.
Although the central system controls all electrics, vacuum pumps, valves etc. ... this control can be handled manually in the facility.



Figure 10. Oven control panel

The operational choices in the control room are the following :

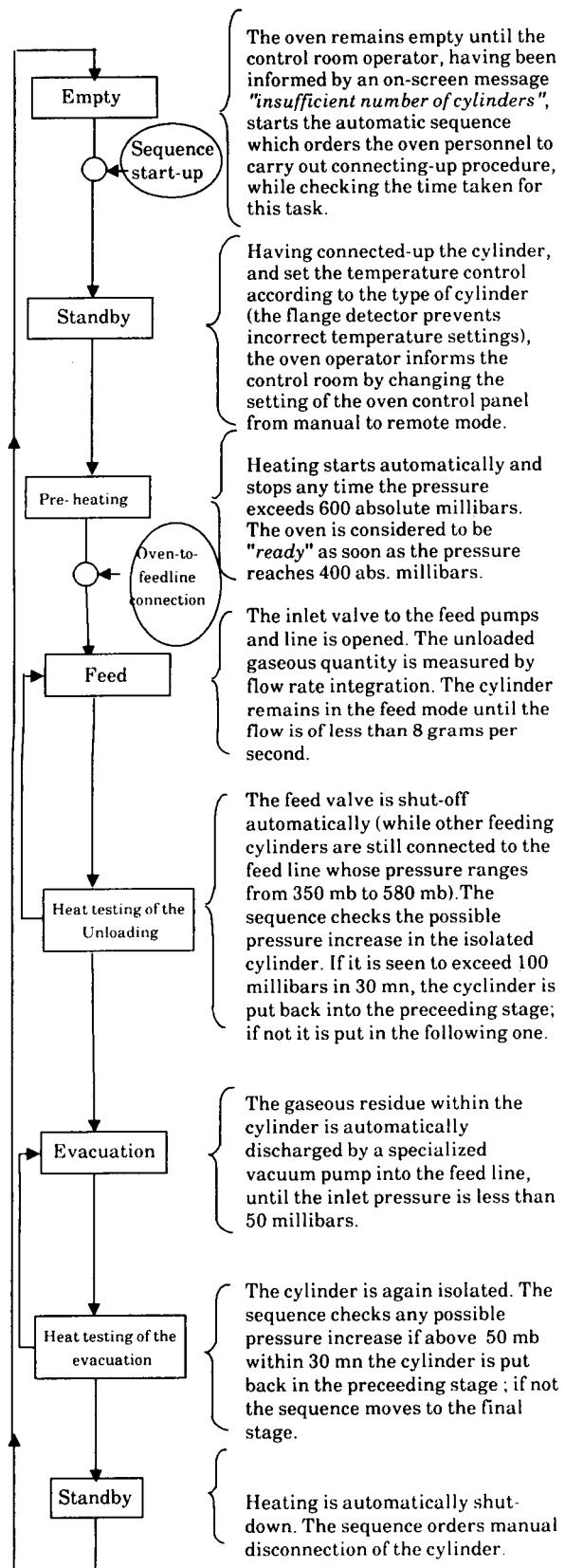
- . Individual control of each piece of equipment.
- . Control of basic sequences concerning pumps, ovens and flow captors.
- . Control of the series of operations which make up the in-oven cycle for any cylinder.
- . Control of the complete management program for the three different feeds F,G,H.

4.2. Computerized operating procedures

Three automatic modes are available :

mode HP	high pressure mode using UF6 liquefaction
mode LPY	low pressure mode using UF6 sublimation from 48 Y cylinders
mode LPG	low pressure mode using UF6 sublimation from 48 G cylinders

The high pressure UF6 liquefaction process (mode HP) is still available but no longer used. The two low pressure UF6 sublimation processes (modes LPY and LPG) are in operation. The different phases of cylinder emptying are computer-controlled from start to finish. Once the controlled sequence is initiated by the control room operator it runs its course without any need of the oven personnel (except for connecting-up and disconnecting). The monitoring of several parameters and alarms (see § 4.3.) allows the computer to verify that all necessary conditions are fulfilled before authorising the sequence to move on from one stage to another. This automatic sequence is described below :



4.3. Monitoring

Several parameters are permanently monitored. They are used by the automatic operating sequence to actuate alarms or to allow the sequence to move from one stage to the following one. The location of the different detectors and their operating ranges are shown in figure 11.

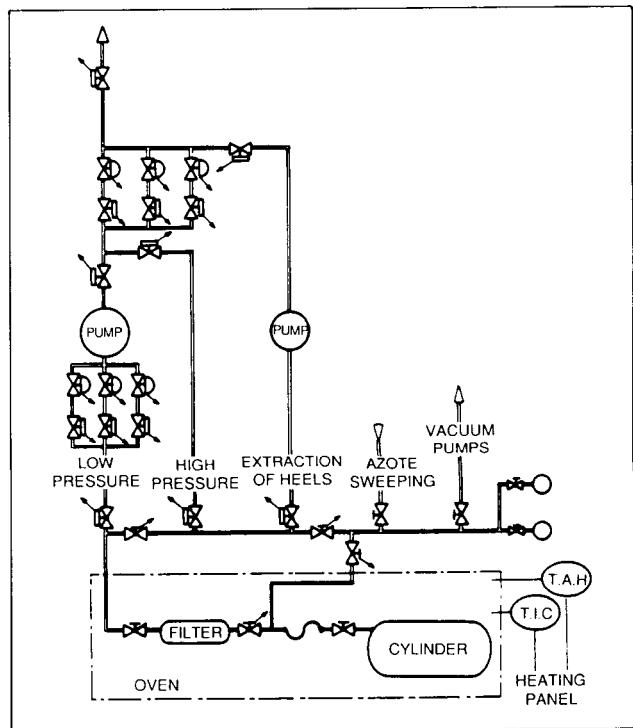


Figure 11. Feed line diagram

The main thresholds and safeguards used by the sequence are listed below:

Automatic Thresholds and Safeguards				
Equipment	Instrument	Permanently monitored threshold		Actuation
Feed cell	Pressure sensor	heating start-up pressure + 40 mb	above	message "cylinder valve closed or obstructed" if not reached after 60 mn of heating
		400 mb	above	message "cylinder ready to feed"
		600 mb	above	stops heating
		9000 mb	above	Automatic disconnection of the feed cell
		end of feeding pressure + 100 mb		Automatic putting back of cylinder in feed mode
		end of evacuation pressure + 50 mb	above	Automatic putting back of cylinder in evacuation mode
	Flow rate sensor	8 g/s (mode LPY) or 5 g/s (mode LPG)	below	Automatic moving from "feed" mode to "heat testing"
		110 g/s	above	message : "high flow rate"
	Oven temperature sensor	109°C (mode LPY) or 79°C (mode LPG)	below	message : "low temperature"
		121°C (mode LPY) or 91°C (mode LPG)	above	message : "high temperature" and automatic termination of the heating
	Flange detector			message : "48 Y or 48G cylinder is connected"
	Timer	several alarms control the sequence timing		message : "overduration"
Feed line	Pressure sensor	350 mb	below	message : "insufficient cylinders in line"
		580 mb	above	message : "Too many cylinders in line"
	Flow rate sensor			Flow rate control
Pumps	Electric power	17.5 KW	above	Pump shut-off
	Outlet pressure sensor	2000 abs.mb	above	Pump shut-off

5 - OPERATING EXPERIENCE

5.1. Process performances

The operating performances of the different feed processes are compared below :

Cylinder model	Feed mode	HP Liquefaction	LP Sublimation	
		48 Y	48 Y	48 G
P E R F O R M A N C E S	Oven temperature	115°C	115°C	85°C
	Pressure abs. mb	1000-4000 Normal range maxi	350-600	350-600
	Average flow rate*	6000	2500	1500
	Net flow rate **	108g/s	55g/s	32g/s
T I M I N G	Connecting-up	90 mn	90 mn	90 mn
	Pre-heating	20 hours	90 mn	90 mn
	Unloading (test included)	31 hours	60 hours	117 hours
	Evacuation (test included)	75 mn	75 mn	75 mn
	Disconnecting	90 mn	90 mn	90 mn

*(Average flow rate) = (amount of UF6 unloaded during feed stage) divided by (feed stage time)

**(Net flow rate) = (total amount of UF6 unloaded during feed and evacuation stages) divided by (time spent by the cylinder in the oven, including feeding, evacuation, connecting and disconnecting times).

The minimum number of ovens and pumps required when using low pressure modes is set out below :

	LP	LP 48Y + 48 G	
Cylinder model	48 Y only	48 Y	48 G
UF6 flow rate g/s	1100	650	300
Number of required ovens	22	13	11
Number of required pumps	3	2	1

Tests have shown that if the cylinder is isolated at any moment during unloading and the heating shut off, the accumulated heat is insufficient to liquefy the remaining UF6 in LPG mode. With the temperature control set to 85°C, the worst case situation is that of a cylinder at the end of unloading and with a low flow rate.

With 150 Kg of UF6 remaining, 1500 mb would be reached only after 9 hours and this for a period of 2 hours.

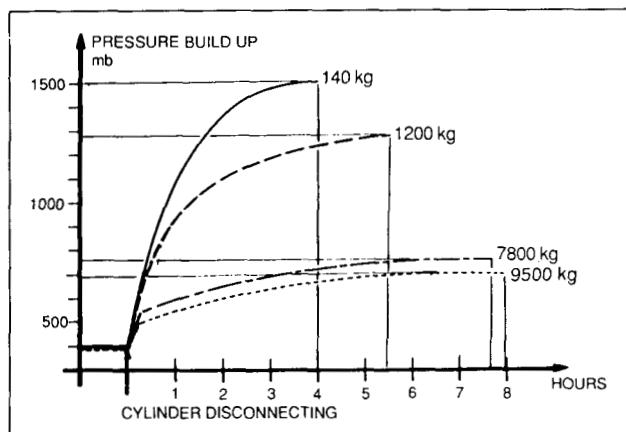


Figure 12. Incidental pressure build-up

Therefore, pressure build-up is sufficiently slow to allow either the reconnection of the cylinder when feeding restarts or the one-off evacuation of a cylinder having a pressure which has built-up too high. In the LPY mode, with the same conditions, except an initial temperature control of 115°C, liquefaction would occur and the pressure would reach 2500 mb (to be compared with the 5000 mb allowed by our specification in that case - see § 2).

A long trial period was necessary to adjust the automatic sequence control parameters in order to make the sequence capable of running from start to finish in any circumstances, and to bring the cylinder from connection to disconnection while allowing its unloading and evacuation in a satisfactory way. End-of-unloading criteria were particularly delicate to adjust. There can be some doubt concerning the residual amount of UF6 within the cylinder, above all when the cylinder tare is unknown.

5.2. Equipment

Filters

No clogging-up of filters has been noticed, the only stripping-down has been for regular inspections (every 3 years) required by the "Service des Mines" (national regulatory body). Cleaning and decontaminating of the filter are carried out outside of the unit and present no particular difficulties. The essential purpose of these filters was to prevent any non-volatile impurities from entering the pressure sensors, the valves and the vacuum pumps which are downstream. In this regard, they have proved to be efficient.

Pumps

One operation limit is to not exceed 110°C at the vacuum pump discharge in order to avoid the decomposition of UF6 into UF4 or other products.

The lowering of pump oil temperature using a thermostatic valve set to 68°C has meant a reduction of 5°C in the temperature of the discharge pipe casing. Since start-up, the number of operating hours for these pumps had amounted to 78000 hours.

By reducing the compression ratio and using an electric power trip, the formation of deposits within the core of the pump has been avoided.

Flow-rate measure

This is currently carried out by membrane pressure-loss gauges (ECA sensor). We are in fact installing thermic flow-rate sensors (SETARAM*) which provide better accuracy.

Leak detection

This is carried out by means of mini ictometers for pollution detection (MIP) which work by accumulating the UO2F2 resulting from the hydrolysis of UF6, on a filter placed in a detection chamber. Recent tests using ionic fume detectors have demonstrated that these latter provide higher sensitivity.

5.3. Lessons learnt from incidents

Cylinder valve obstruction caused by crystallized UF6 or by UO2F2, has occurred several times :

- Once there was a case of valve jamming but finally the valve was opened and the flow-rate was found to be normal.
- In another case a valve was opened but there was no resulting flow. The obstruction was inside the needle seating of the valve. Measured pressure build-up was abnormal after 30 minutes heating and heating was immediately shut down.
- In a third case, the flow was noticed to be extremely low after valve opening (obstruction caused by porous UO2F2). This case is the most delicate as it is necessary to interpret the difference between the nominal flow curve and the actual one in order to shut down as quickly as possible if necessary to avoid an unknown pressure increase inside the cylinder.

6 - CONCLUSIONS

The feed process using sublimation has proved to be very flexible allowing flow rates of up to 1100 g/s. All operating procedures are automatically controlled except for the connecting and disconnecting of cylinders which are carried out manually.

There have been no operating incidents from this automatic procedure.

As we have seen, EURODIF'S GEORGES BESSE plant has the capability to feed up to 35000 tons of Uranium hexafluoride. Therefore, it is one of the world leaders in enrichment capacities and will remain up to the beginning of the next century.

*SETARAM : Société d'Etudes d'automatisation, de régulation et d'appareils de mesures.

SAFE HEATING OF 48G CYLINDERS CONTAINING UF₆

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ABSTRACT

The gaseous diffusion plants produce enriched uranium to be used as fuel in nuclear reactors. In doing so, large quantities of 0.2% U₂₃₅ tails are produced. This material is in the form of solid UF₆ and is in long-term storage in 48G cylinders at the three plants.

The 48G cylinders is designed and, is designated in ORO 651 Rev. 5, to be a low cost container "for tails storage only". Recently a need has developed to use some of these cylinders for short-term storage of toll enrichment normal assay UF₆ feed and a program has developed to utilize the tails in another process.

This report is intended to provide the basis for developing safe procedures to heat the 48G cylinders in steam-heated autoclaves to remove the UF₆. The effect of the variables of cylinder volume, net weight of UF₆, cold cylinder pressure, and autoclave temperature are considered and limits on these variables are stated.

To support these conclusion, the characteristics of UF₆ are provided, the cylinder is described, and calculations of cylinders internal pressures are presented.

UF₆ CHARACTERISTICS

UF₆ is a solid compound at room temperature, but can be maintained in the gas phase under controlled temperature and pressure conditions. In Figure 1, the phase relationship of pure UF₆ is depicted as a function of pressure, expressed in psia, versus temperature in degrees Fahrenheit. The triple point, the only combination of pressure and temperature where all three phases exist simultaneously, exists at a pressure of 22 psia and a temperature of 147°F. If either, or both, pressure and temperature are lower than these values, the UF₆ will be a solid and/or gas. When

the pressure and temperature are increased above the triple point conditions, only liquid or gas will exist.

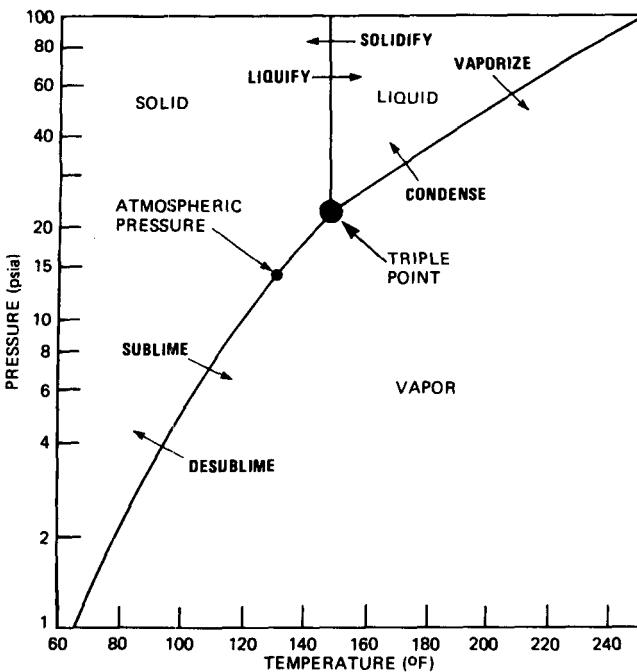


Figure 1. Phase diagram of UF₆.

UF₆ can be handled safety in essentially the same manner as any other corrosive and/or toxic chemical by employing procedures developed to accommodate the characteristics of the material. Illustrated in Figure 2 is a sealed glass vial that has been evacuated to remove the air, partially filled with liquid UF₆, and allowed to cool. The illustration is provided to help the reader visualize the conditions inside a UF₆ cylinder. The photograph was taken where the ambient temperature was 100°F. It shows off-white colored solid UF₆ crystals surrounded by colorless UF₆ gas. If a pressure gauge were connected to the vial, it would indicate a pressure of 5 psia. There is only gaseous and solid UF₆ in the tube. If the tube is heated, some of the crystals will disappear as they sublime to the gas phase, causing an increase in pressure. Appropriate instrumentation would indicate that the heat required to vaporize the solid UF₆, the heat of sublimation, is 58.2 BTU/lb. If the tube is heated to a temperature of 147°F, the pressure will be 22 psia and drops of colorless liquid will appear. If heating is continued, the pressure will remain at 22 psia until all of the solid is

melted in a phase change, during which the heat of fusion is absorbed by the solid. As heating of the vial continues, the heat causes a rise in temperature of the liquid and, as the temperature increases, the liquid vaporizes causing an increase of pressure.

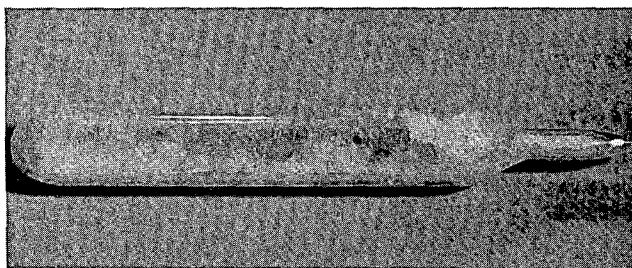


Figure 2. Glass Vial Containing UF₆.

Exactly the same changes occur in all cylinders which contain UF₆. Regardless of its isotopic content, pure UF₆ will follow the phase diagram. However, if a cylinder contains impurities in the form of traces of other gases, the pressure encountered will be higher by a factor that is directly related to the phase relationships of the impurities. The most common impurities encountered are hydrogen fluoride (HF), Freon-114, and air. The presence of these impurities can be detected by measuring the gas-phase pressure of an unheated cylinder and comparing it to the UF₆ phase diagram. This vacuum check at ambient temperature is frequently referred to as a "cold-pressure check". Impurities can cause the pressure to be significantly higher than the vapor pressure of 5 psia at 100°F ambient temperature, but will be far below the design pressure rating for the cylinder. Because these impurities have higher vapor pressures than UF₆, they can be removed by evacuating some of the gas contents from the cold cylinder. This evacuation procedure is analogous to a cold distillation in which the high vapor pressure components of the mixture are vaporized first. This is commonly referred to as "cold burping". Some UF₆ is removed during this process, but the quantity is in the order of a few pounds per 10-ton or 14-ton cylinder. The burp gas is drawn off through cold traps designed to collect the UF₆ and pass the impurities.

The important physical characteristic that must be considered when liquid UF₆ is placed in cylinders, is the change in liquid density as a function of temperature. Figure 3 shows this relationship. It is this factor which can create an overfilling if not properly taken into account. In the gaseous diffusion plant, cylinders are filled with liquid UF₆ at a temperature of 160°F, with the density of the material being 224.5 lb/ft³. To illustrate what could happen, consider that a cylinder with an internal volume of 139 ft³ is filled to 97 percent of its volume capacity with liquid UF₆ at a temperature of 160°F. The cylinder would accept 30,270 lbs. in this case. However, if this cylinder is then heated to some temperature about 160°F, the 3 percent remaining free volume or ullage will gradually decrease because at higher temperatures each pound of liquid UF₆ occupies more volume. In this example,

when the contents reach a temperature of 194°F, the cylinder will be completely full. Any further heating will cause the confined liquid to hydraulically deform and rupture the cylinder.

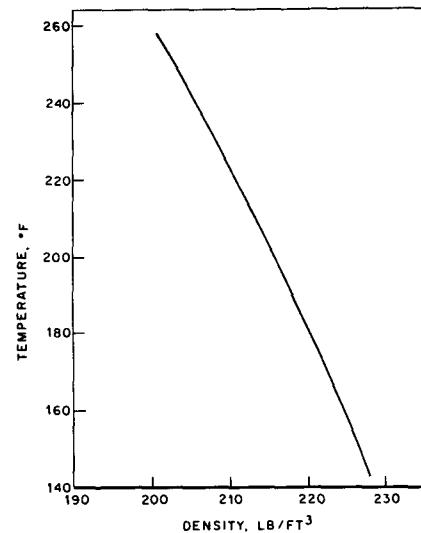


Figure 3. Density of liquid UF₆.

This is a well understood phenomenon, and hydraulic rupture is prevented by establishing cylinder fill limits based on the UF₆ density at 250°F for all cylinders, except the 48G where 235°F is the basis when filled with tails. In DOE owned facilities, cylinders are heated in autoclaves with a controlled steam pressure of approximately 8 psig to guarantee that the contents will not exceed a temperature of 235°F. In addition to taking the maximum liquid temperature into account, the quantity of UF₆ is limited to assure at least a 3% ullage when the full cylinder is at its design temperature. This gas phase volume is necessary to avoid the possibility of exceeding the cylinder design pressure due to the presence of high-vapor pressure containments. Adherence to these safe heating conditions is assured by (1) publishing the cylinder fill limits, (2) requiring the cylinder manufacturer to measure the cylinder volume by weighing it full of water and stamping this water capacity on the cylinder nameplate, (3) maintaining accurate scales to determine the net contents of a cylinder, (4) operating the withdrawal stations to yield high purity UF₆ (5) controlling the maximum steam heating pressure, and (6) utilizing the cold cylinder vapor pressure measurement to verify the amount of volatile impurities present before heating the cylinders.

48G CYLINDER CHARACTERISTICS

In the past forty years, a family of different-size cylinders has been developed to safely contain UF₆. These approved cylinders are described in Report ORO-651, Rev. 5, Uranium Hexafluoride: Handling Procedures and Container Descriptions and the American National Standard for Packaging of Uranium Hexafluoride for Transport, ANSI, N14.1-1987. The 48G cylinder was designed to be a minimum cost, large-capacity container for long-term storage of tails assay UF₆ at the gaseous diffusion plant.

Figures 4 and 5 are presented to show general data and some of the design details of the cylinder.

The fundamental difference between this cylinder and the other large UF₆ cylinders is the shell thickness. They are fabricated from 5/16" thick steel and the others have 5/8" thick steel.

The service pressure or maximum operating pressure for the 48G cylinder is 100 psig and the hydrotest

pressure is 200 psig. It has a certified minimum of volume of 139 ft³.

As of August 1983, there were about 15,500 of these cylinders filled with 0.2 percent U₂₃₅ tails stored at the three diffusion plants. Each cylinder costs about \$2000, and because so many are used, the intent is to fill them as full as possible. This is done by using a smaller ullage factor than is used for any other UF₆ cylinder.

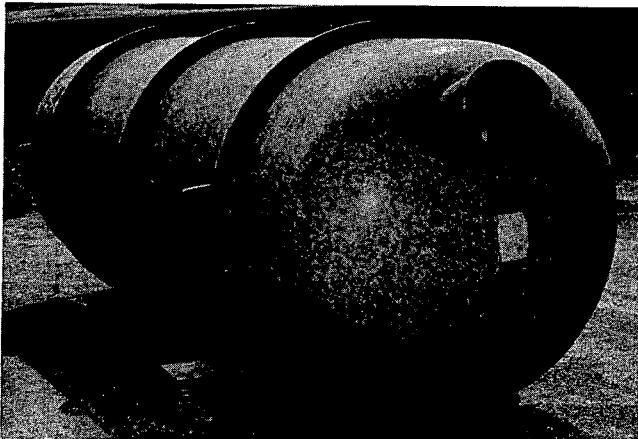


Figure 4. UF_6 cylinder Model 48G.

GENERAL DATA

Nominal Diameter	48 in.
Nominal Length	146 in.
Nominal Wall Thickness	5/16 in.
Nominal Tare Weight	2,600 lb (1,179 kg)
Maximum Net Weight	28,000 (12,701 kg)*
Nominal Gross Weight	30,600 lb (13,880 kg)
Minimum Volume	139 ft ³ (3.94 m ³)
Basic Material of Construction	Steel**
Service Pressure	100 psig
Hydrostatic Test Pressure	200 psig
Isotopic Content Limit	1% U-235

Valve Used - 1-in. Valve.
*Based on 235° F (113° C)

**Steel specification changed from A-285 to A-516 for cylinders ordered after 1978.

NOTE: For tails storage only. Cylinders with serial numbers below 111821 do not have certified volumes. An earlier design was designated Model 'OM'.

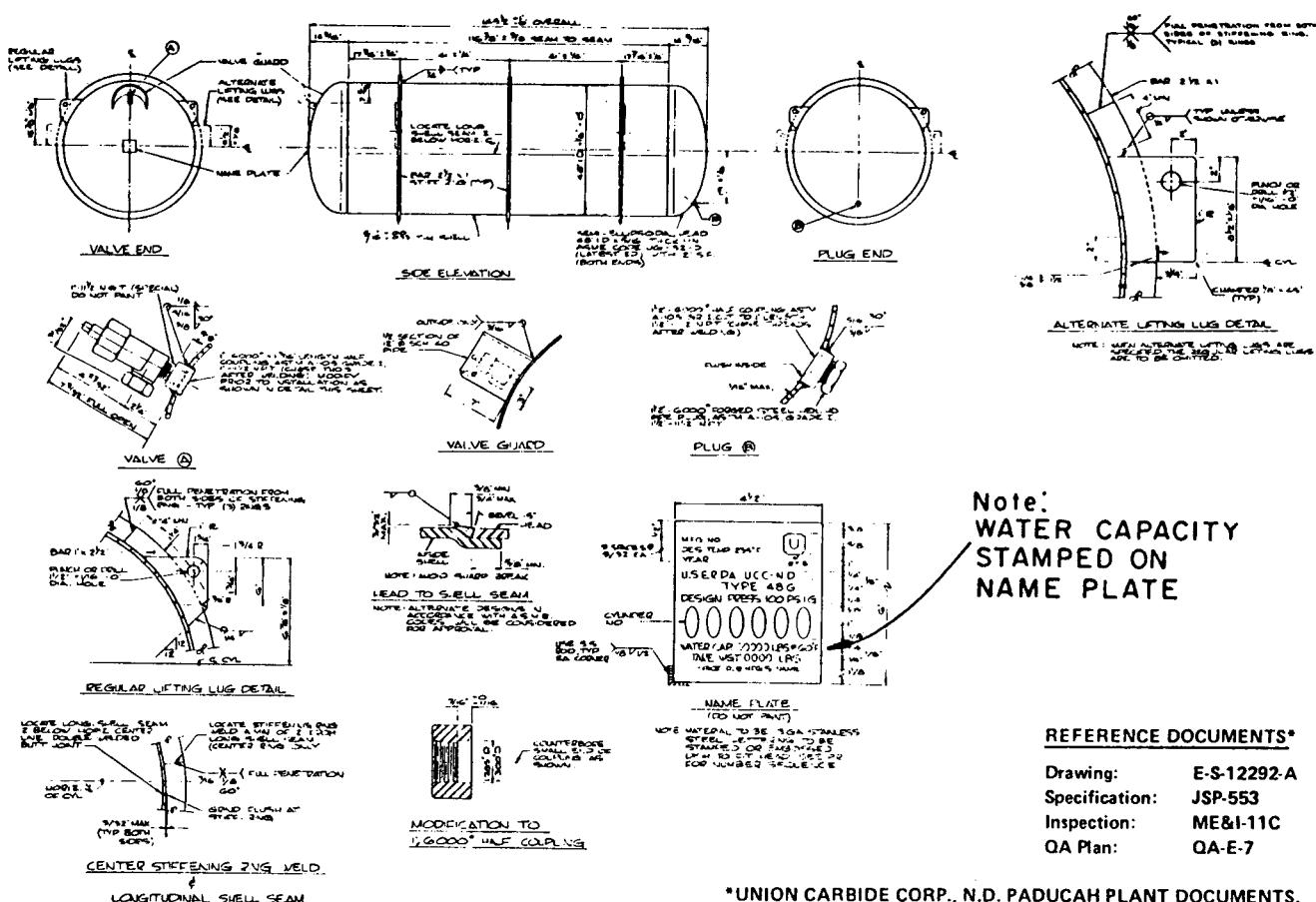


Figure 5. UF₆ cylinder Model 48G design details.

REFERENCE DOCUMENTS*

Drawing: E-S-12292-A
Specification: JSP-553
Inspection: ME&I-11C
QA Plan: QA E-3

*UNION CARBIDE CORP. N.D. PARUCHAH PLANT DOCUMENTS

The 48G cylinder maximum fill limit is 28,000 lb. UF₆. This is the quantity of tails assay UF₆ which when heated to 235°F will leave a 3 percent free volume in a cylinder whose certified minimum volume is 139 ft³. The contents of the ullage is assumed to be composed of UF₆ gas and a small amount of air. The partial pressure of UF₆ gas will be the vapor pressure for the temperature of the liquid UF₆ which has been put in the cylinder.

The partial pressure of air will be the result of compressing the initial quantity of air in the empty cylinder into the final ullage.

Each year the Paducah Gaseous Diffusion Plant has the responsibility to procure new 48G cylinders for the three diffusion plants. Different manufacturers supply them, but since they are all built to the same drawings and specifications, they are all nearly identical. One of the important features of the cylinder is its certified minimum volume of 139 ft³; this is the value that determines the UF₆ fill limit. Beginning in 1972, the minimum volume for all cylinders above serial number 111821 has been certified by weighing the cylinder full of 60°F water and stamping the water capacity weight on the stainless steel cylinder nameplate. Because of the importance of this weight, the scale used for the determination is accurate to +0.1 percent. To assure that the scale maintains its accuracy, it is checked frequently with a check weight, which is a 48G cylinder full of water that has been weighed on the Paducah uranium accountability scales that are accurate to +0.001 percent.

If a cylinder has a volume of exactly 139 ft³, its water capacity at 60°F would be 8673 lb.

At the K-25 Plant a random check of 46 of the 48G cylinders, made by three manufacturers over a period of 13 years, showed the mean water weight to be 8980 lbs. with a low of 8917 lb. and a high of 9041 lb. These weights correspond to an average volume of 143.9 ft³, with a low of 142.9 ft³ and a high of 144.9 ft³. The sample included both new empty cylinders and ones that had been filled for years.

CYLINDER PRESSURE CALCULATIONS - 235°F

The Normal Operating Temperature Limit for UF₆ autoclaves, specified in the Operational Safety Requirements (OSR) for the three diffusion plants and the Feed Material Production Center is 235°F. Therefore, the pressure developed when a full cylinder is heated to 235°F needs to be examined.

Assume that a cylinder is stamped with a water capacity of 8950 lb., contains 28,000 lb. UF₆, and its cold pressure is 5 psia.

$$\text{Ideal Gas Law: } \frac{P_1 V_1}{T_1} = \frac{P_2 V_2}{T_2}$$

Where:

P₁ = cylinder cold pressure
 V₁ = cold cylinder free volume
 T₁ = cold cylinder temperature °Rankin
 P₂ = cylinder hot pressure
 V₂ = hot cylinder free volume
 T₂ = hot cylinder temperature °Rankin

P₂ is the pressure of interest so the equation becomes:

$$P_2 = \frac{P_1 V_1 T_2}{T_1 V_2}$$

According to Dalton's Law the total pressure of a gas is equal to the sum of the partial pressures of the components, therefore, if the cylinder has a cold pressure of 5 psia at 68°F when the vapor pressure of UF₆ is 1.5 psia, then the pressure of the non-condensable gas is

$$P_1 = 5 - 1.5 = 3.5 \text{ psia}$$

Total volume of

$$\text{cyl.} = \frac{8950 \text{ lbs water}}{62.4 \text{ lb/ft}^3 \text{ water}} = 143.4 \text{ ft}^3$$

Density of solid UF₆ at 68°F is 317.8 lbs/ft³, therefore

$$\frac{28,000 \text{ lbs UF}_6}{317.8 \text{ lbs/ft}^3} = 88.1 \text{ ft}^3 \text{ of solid UF}_6 @ 68^\circ\text{F}$$

$$V_1 = 143.4 - 88.1 = 55.3 \text{ ft}^3$$

Density of liquid UF₆ at 235°F is 207.1 lbs/ft³, therefore

$$\frac{28,000 \text{ lbs UF}_6}{207.1 \text{ lb/ft}^3} = 135.2 \text{ ft}^3 \text{ of liquid UF}_6 @ 235^\circ\text{F}$$

$$V_2 = 143.4 - 135.2 = 8.2 \text{ ft}^3$$

$$T_1 = 460 + 68^\circ\text{F} = 528^\circ\text{R}$$

$$T_2 = 460 + 235^\circ\text{F} = 695^\circ\text{R}$$

Substituting these values in the equation:

$$P_2 = \frac{(3.5)(55.3)(695)}{(528)(8.2)}$$

$$= 31.1 \text{ psia of air}$$

$$= +79.0 \text{ psia vapor press of UF}_6 @ 235^\circ\text{F}$$

110.1 psia total pressure in the cylinder

Similar calculations have been made for different cylinder volumes and are shown in Figure 6. Note that if the cold pressure is 5 psia, the pressure in the cylinder at 235°F is lower than the cylinder service pressure for water capacities greater than 8870 lb. This would be a minimum acceptable water capacity to use as a guide for feeding 48G cylinders containing 28,000 lbs of UF₆. However, considering the inaccuracies of the calculation, the inaccuracies of the scales used to determine the water capacity, and the desire to operate slightly below the cylinder service pressure, a water capacity of 8950 lb. should be set as the minimum acceptable.

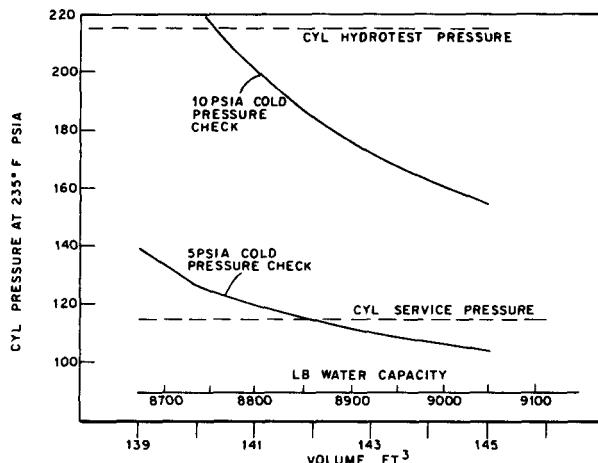


Figure 6. UF_6 cylinder Model 48G, 28,000 lb. at 235 °F.

Also shown on Figure 6 is the cylinder pressure at 235°F if a 10 psia cold pressure check were used. No cylinder volume provides an acceptable cylinder pressure. The 5 psia or less, cold pressure check must be the rule for heating cylinders to 235°F.

The small random sample of cylinders examined at K-25, had standard deviation of 27.9 lb. around the mean water weight of 8980 lb. Assuming the standard deviation includes values within ± 34 percent of the mean, eliminating from consideration any cylinder with less than 8950 lb. water capacity from the low end of the distribution will rule out only about 16 percent of the available cylinders from heating in this manner.

Similar calculations have been made to determine a minimum safe water capacity for the case of the 48G cylinder filled to 26840 lbs. of normal assay UF_6 and heated to 235°F. As shown in Figure 7, as long as the 5 psia cold pressure check is observed, any water capacity greater than 8680 lbs. is acceptable. Again, the 10 psia cold pressure check yields pressures above the cylinder service pressure.

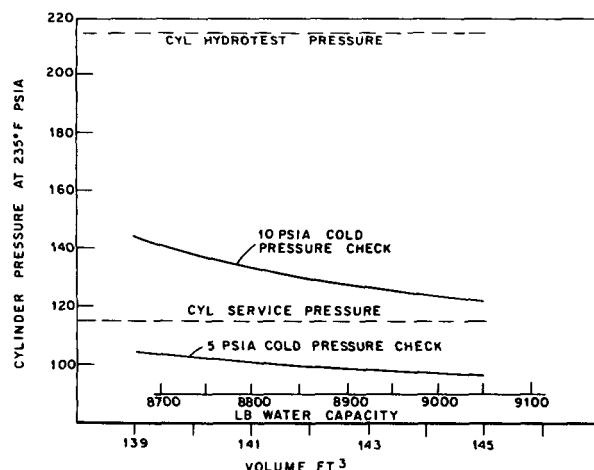


Figure 7. UF_6 cylinder Model 48G, 26,840 lb. at 235 °F.

CYLINDER PRESSURE CALCULATIONS - 200°F

The 235°F calculations clearly show that the cold pressure check of 5 psia must be observed to avoid

exceeding the cylinder service pressure. In the case of sampling a cylinder of normal assay feed for UF_6 purity, where removing gas to observe the 5 psia limit would affect the sample analysis, a lower temperature heating regime should be followed. Three of the four DOE plants have magnetically-connected thermocouples on the outer surface of the cylinder. This temperature indication can be used to control the steam input to the autoclave to control the cylinder temperature. Assuming the control is set to control at 200°F, Figures 8 and 9 show the hot full cylinder pressures which result. With either quantity of UF_6 , the usual 10 psia cold pressure check yields acceptable results.

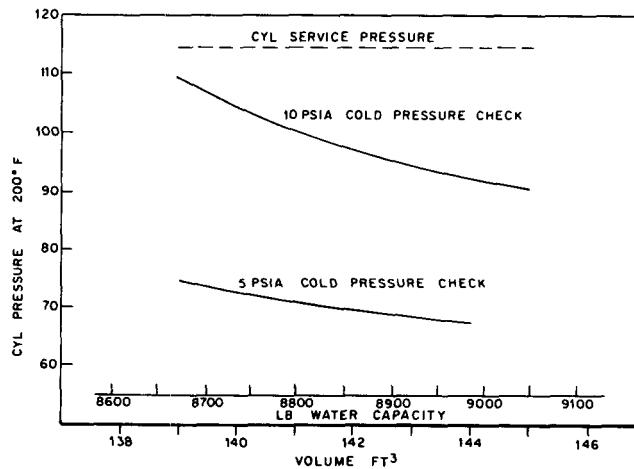


Figure 8. UF_6 cylinder Model 48G, 28,000 lb. at 200 °F.

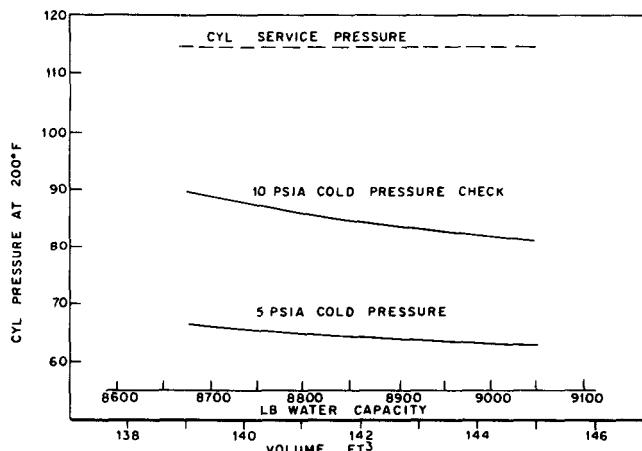


Figure 9. UF_6 cylinder Model 48G, 26,840 lb. at 200 °F.

OTHER GASES IN THE CYLINDER

These calculations have considered the effect of dry air as the other gas in addition to UF_6 in the cylinder. The UF_6 tail stream is taken from the bottom of the gaseous diffusion cascades and contains no HF or Freon 114 because of the separating characteristics of the cascade. When the cylinders are filled, the only impurity is the small amount of dry air that was in the empty cylinder before it was filled.

However, it is conceivable that a cylinder in storage could have its valve seat leak, permitting the free volume to rise to atmospheric pressure with wet air, thereby producing some HF from the reaction of water with UF₆. The result of this was calculated in the following manner:

Assuming the 28,000 lb. of solid UF₆ to be at a temperature of 68°F with a density of 317.8 lb/ft³, there will be 55.3 ft³ of free volume in the cylinder.

The vapor pressure of UF₆ at 68°F is 1.5 psia. Therefore, 14.7 psia - 1.5 psia = 13.2 psia = partial pressure of air in the ullage.

This is equivalent to

$$\frac{13.2 \text{ psia}}{14.7 \text{ psia}} \times 55.3 \text{ ft}^3 = 49.7 \text{ Standard Cubic Feed (SCF) of Air}$$

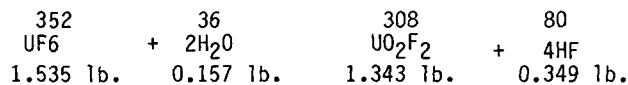
One pound of saturated air at 68°F occupies 13.613 SCF. Thus

$$\frac{49.7}{13.613} = 3.65 \text{ lbs of air would exist in the cylinder,}$$

Assuming the leakage occurred when the air temperature was 100°F and the relative humidity was 100 percent, there would be 0.043 lb. H₂O/lb air.

3.65 x 0.043 = 0.157 lb. H₂O available for reaction with UF₆ according to the following equation.

molecular weights



The 0.349 lb. of HF produced by the reaction can be expressed:

$$\frac{0.349 \text{ lb. HF}}{20 \text{ Mol wt. HF}} = 0.0175 \text{ mol HF}$$

$$0.0175 \times 359 \text{ (lb. molecular volume)} = 6.26 \text{ SCF}$$

This amount of HF in the 55.3 ft³ ullage of the cylinder is

$$\frac{6.26 \text{ SCF}}{55.3 \text{ SCF}} = 11.3\% \text{ of the gas mixture}$$

Since the total pressure in the ullage is 14.7 psia, of which 11.3% is HF, then 1.66 psia is the partial pressure of HF. The vapor pressure of pure HF at 68°F is 14.9 psia so all the HF in the ullage will stay in the gas phase and not condense to liquid. Therefore, when the cylinder is heated

after being evacuated to 5 psia leaving only 0.12 lb of HF, the increasing pressure of this small quantity of HF gas will not cause the final pressure to be any different than if the ullage had been full of air only.

HYDRAULIC FORCES

Hydraulic forces created by expansion of liquid UF₆ cannot rupture a 8950 lb. water capacity cylinder containing 28000 lbs. of UF₆ when it is heated to 235°F in an autoclave. The design of the autoclave utilizes a steam-pressure controller to limit the steam pressure to 8 psig (235°F), and cylinder skin temperature monitors are installed to automatically cut off the steam if the cylinder wall temperature exceeds 235°F. A cylinder with volume of 143.4 ft³, and filled with 28,000 lb. UF₆, would have to be heated to 279°F (steam pressure of 33 psig) to completely fill it with liquid UF₆ and establish the potential for hydraulic rupture.

CONCLUSIONS

Adhering to the following provisions will assure safe heating of 48G cylinders.

1. No cylinder with a serial number lower than 11821 should be heated.
2. No cylinder with greater than 28,000 lb. of UF₆ should be heated.
3. No cylinder should be heated to 235°F with a water capacity of less than:
 - a. 8950 lb. when filled to no more than 28,000 lb. UF₆ or
 - b. 8680 lb. when filled to no more than 26,840 lb. UF₆.
4. The ullage of the cylinder should be evacuated to 5 psia or less before heating to 235°F.
5. The steam pressure to the autoclave should not exceed 8 psig for 235°F service.
6. The cylinder skin temperature thermocouples in the autoclave should be set to shut off the steam at 235°F.
7. The cylinder internal pressure measuring instruments should be set to shut off the steam no higher than 114.7 psig. (100 psig)
8. When sampling a cylinder, where the cold pressure cannot be reduced to 5 psia, the cylinder skin temperature thermocouples should be set to control the cylinder temperature at 200°F by adjusting the steam flow.

UF₆-RELEASE IN A GERMAN FUEL FABRICATION PLANT
 - SEQUENCE AND CONSEQUENCES -

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ABSTRACT

In April 1987 a UF₆ release occurred in the conversion facility of the Reaktor-Brennelement Union at Hanau, F.R. Germany. The incident sequence started with a small leakage at a stuffing box of the main UF₆ valve of an evaporation autoclave. The operators stopped the normal evaporation process and closed the main UF₆ valve and the UF₆ cylinder valve. Obviously the remotely driven UF₆ cylinder valve was not closed totally and was not checked by hands. So a release occurred out of the exhausting device. Due to the water in the evacuation pump a plug of UO₂F₂ was built up in the exhausting line near the pump and then the pressure increased until the flexible tube broke open. This major release was stopped after some minutes by operators wearing respirators. The released UF₆ remained totally within the building and no injury to personal health was observed although the whole floor and all components in the building were covered with UO₂F₂. Details of the installations, the incident sequence, the radiological impact as well as the consequences, the improvement in the technical equipment and the operation manual are described.

PROCESS AND EQUIPMENT

In the fuel fabrication plant of the Reaktor-Brennelement Union (RBU) at Hanau F.R. Germany uranium dioxide powder is produced by the AUC-conversion process /1/. The gaseous UF₆ is lead into a aqueous fluid simultaneous with CO₂ and NH₃. The resulting ammonium uranyl carbonate (AUC) is precipitated and filtered and later on treated by thermal decomposition and reduction in a fluidized bed reactor (fig. 1).

For the evaporation of the low enriched uranium hexafluoride, which is in 30" cylinders, steam heated autoclaves are used. The precipitator is a slab vessel with a circulating loop. The

process gases are fed into the aqueous fluid in nozzles within the loop. The off gas of the vessel is lead to a jet scrubber and further on to a spray scrubber and the filter system.

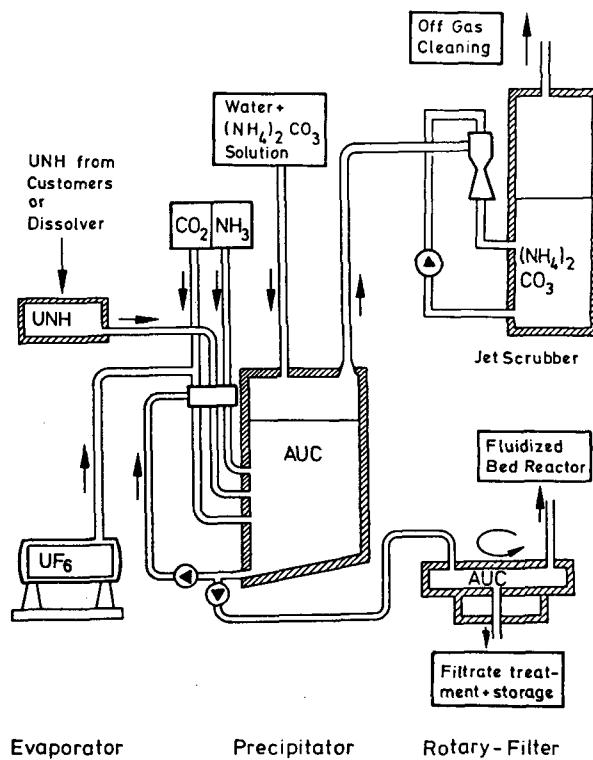


Fig. 1: AUC-Precipitation

The UF_6 cylinder valve can be driven remotely. The pigtail within the autoclave consists of stainless steel. The UF_6 line from the autoclave to the precipitation vessel is electrically heated. The main closing valve for safety shut down is located directly above the autoclave (fig. 2). Between this valve and the controlling valve the exhausting device and the CO_2 flooding system are connected, which are normally closed by a pneumatic valve. After branching of exhausting line and CO_2 -line there are hand valves. Near the T-piece a pressure gauge is installed.

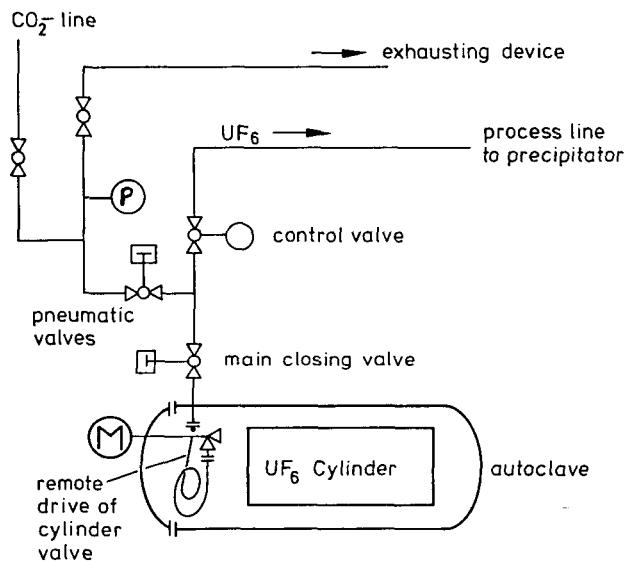


Fig. 2: Evaporating Equipment

The exhausting device consists of a liquid seal pump with a fluid loop connected to the pool of the jet scrubber (fig. 3). To protect the pump against cavitation there is a vacuum breaking by pass. The exhausting line is partially constructed with a flexible tube, since the plant was in a back-fitting procedure concerning the exhausting equipment.

In normal operating conditions the vacuum system is used to exhaust small UF_6 discharges especially during cylinder changing. The CO_2 gas system is used for the leakage tests before heating the UF_6 cylinders and for emptying the gaseous residues. For this purpose CO_2 is fed in the almost empty cylinder and it works as a delivery gas. All these operations in normal conditions are done according to a detailed check list.

INCIDENT SEQUENCE

On April 27th 1987 at about 11 p.m. there was a heated and almost full cylinder in the autoclave. The batch was started 15 minutes ago.

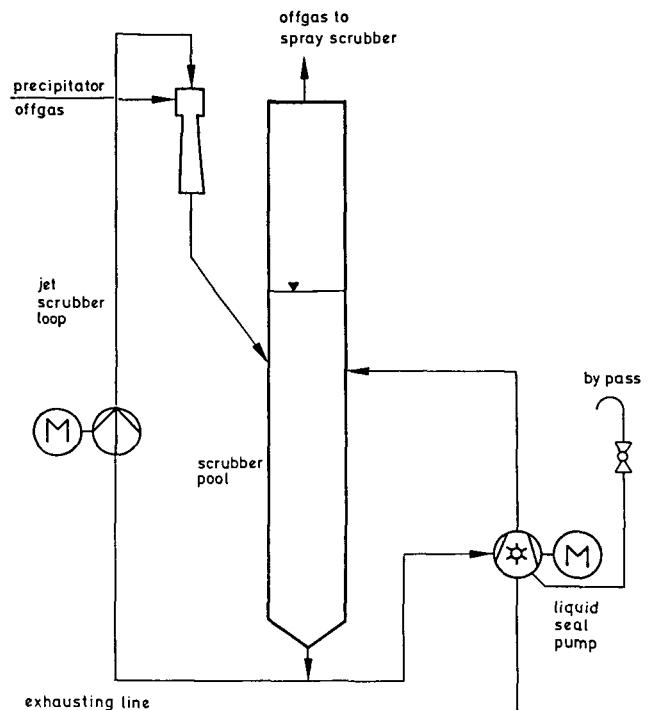


Fig. 3: Jet Scrubber and Exhausting Pump

A small leakage at the stuffing box of the main UF_6 closing valve appeared. The operators stopped the normal evaporation process and closed the UF_6 cylinder valve and the main valve. In order to repair the leaky valve the UF_6 process line had to be evacuated by the exhausting device with the liquid seal pump. Therefore the hand valve was opened and the under pressure was checked at the pressure gauge. Now the two pneumatic valves were opened. Unexpectedly a pressure increase was observed. When the operators were looking for the reason of this pressure rise, an evacuation alarm came and they had to leave the building. The UF_6 released into the hall out of the bypass line of the pump. Since the scrubber is in another room, the operators at the evaporator could not see the release and could not realize the connection between their manipulation and the alarm. The facility remained in this condition with valves opened.

Obviously the remotely driven UF_6 cylinder valve was not closed totally and was not checked by hands. The UF_6 streamed through the exhausting line to the liquid seal pump into the scrubber pool. Due to the water in the pump plugs of UO_2F_2 were built up, first in the small bypass and then in the exhausting tube. These plugs of about 30 to 50 cm length were found two days later during the search for the incident reasons and sequence.

Due to the plug the pressure increased in the line until the flexible tube broke open. This second major release occurred 3 minutes after the alarm near the door between two conversion halls. The time was recorded by the UF_6 warning devices and little later by the fire detectors at the opposite end of the hall. About 10 minutes after alarm two operators wearing full protection and respirators entered the hall and closed all valves by hand and stopped the release. They found the halls totally filled with UO_2F_2 fog. The distance of sight was only 10 cm.

After the incident the floor and the installations of the process halls were found all covered with a UO_2F_2 layer. Investigating the equipment for the incident reasons about 1 kg uranium was found in the UO_2F_2 plugs and in UF_6 plugs within the pigtail and the valves. The cylinder valve and all valves were found closed, but the solid UF_6 found in the normally heated part of the UF_6 line is a hint that the cylinder valve was still open during cool down of the cylinder. The amount of released UF_6 , estimated by weighing the cooled cylinder, was about 50 kg. Of this amount about 10 kg were absorbed in the scrubber.

RADIOLOGICAL CONSEQUENCES

Due to the early evacuation alarm caused by the first small release at the exhausting pump the whole shift had left the hall before the major release occurred.

One hour after the incident the ventilating system was closed because of activity alarm in the off gas. The monitor in the air outlet duct showed an increase to 15900 Bq α -counts per minute. The first evaluation of the survey filters resulted in a activity of 5.9 Bq/m³, so the released activity due to the incident was estimated to 7.95 MBq. In comparison the limit for gaseous effluents is 22.2 MBq per month. The release to the environment was clearly below the permitted value.

Within the building the first measurements of the activity in the air were conducted at 7 a.m. in the morning of the next day. The air in the conversion hall had 30 Bq/m³ α -radiation, in the adjacent rooms only 0.4 Bq/m³ respectively 0.72 Bq/m³ were observed.

In the following hours the main filter elements of the air cleaning center were replaced. The ventilation system returned to operation at 3:30 in the afternoon. The access to the process rooms without respirators or masks was released by the protection survey at 6 p.m., since only 0.3 Bq/m³ in the air were measured. After that the staff began to decontaminate the hall and the installations.

The operating personnel in the chemical process who had left the hall after the evacuation alarm was checked for radioprotection and medical reasons. All of the 23 persons had no symptoms of HF etching. Nevertheless their urine probes were investigated to estimate the uranium incorporation. All of them were

far below the permissible limit. The next day all members of the night shift were investigated once more by the plant physician and no health injury could be observed.

TECHNICAL CONSEQUENCES

Several lessons learned from the mishap concerning the technical installations and the operations have to be mentioned. There are some improvements in the equipment:

- The bypass line of the liquid seal pump has been removed. So the vacuum breaking function to avoid cavitation is suppressed, but that is tolerable in normal operating conditions.
- The planned equipment for exhausting with heated pipes of stainless steel and pumps related to each autoclave has been installed immediately.
- The control of the main closing valve at the autoclave and the pneumatic valve at the exhausting line is improved so that even in the case of repair the emergency shut down functions will guarantee the safe closure.
- The control of the remote drive for the UF_6 cylinder valve is improved so that the imperfect closure of the valve is more unlikely.

Also in the operating manual some details are improved:

- The closure of the cylinder valve has to be checked by hands and by a tightness proof with over and under pressure.
- The repair of the main closing valve at the autoclave or of leaky components within the autoclave will not be conducted until the cylinder has been cooled down.
- The autoclave will not be opened for interventions while a heated cylinder is inside, excepted for a quicker cool down without any manipulations.
- The check list for the normal operation with changing of UF_6 cylinders is enhanced so that the condition of interventions during evaporation is included.

CONCLUSIONS AND SAFETY PRINCIPLES

The technical equipment and the operating procedures are suitable for the safe handling and processing of UF_6 and for plant maintenance. The deficiencies which gave the possibility of the mishap are clearly recognized and the lessons learned lead to some improvements especially in the operators manual and know how.

The principles of the german safety requirements /2/

- autoclaves as a second containment
- building or separate rooms as third containment

for the inclusion of UF_6 during handling in liquid phase or as gas with overpressure secured that no radiological impact to the environment occurred. Even in repair conditions the requirement of a second containment for handling liquid UF_6 will be respected consequently in the future.

The use of liquid seal pumps connected with a scrubber pool for the vacuum generation in the exhausting device seems to fit emergency requirements, since the self sealing behaviour by UO_2F_2 is an advantage if a large amount of UF_6 is released into the device.

To enhance the safety of UF_6 evaporation it is considered to improve the autoclaves with a closed loop for direct steam heating, with a possibility of wall cooling and with separated evaporator rooms. So the multiple containment concept as it is suggested in the safety requirements can be performed in all normal and abnormal conditions.

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EFFICIENT AND SAFE HOT AIR HEATING OF SIZE 5A/B UF₆ CYLINDERS

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ABSTRACT

An efficient and safe means for unloading size 5 VHE uranium hexafluoride (UF₆) cylinders using hot air has been developed and tested at the Savannah River Plant (SRP). Hot air impinges directly on the cylinder through hundreds of small holes close to the cylinder surface. A cylinder of UF₆ can be unloaded in about 10 hours at an air temperature of 100°C. The forced convection heating system replaced a natural convection heater in which the UF₆ cylinder was placed in the center of an electrical resistance-heated 6-in. pipe. A comparison of the two types of heating systems is presented. The overall heat transfer coefficient using forced convection is 11.5 Btu/(hr)(ft²)(°F) as compared to 3.9 Btu/(hr)(ft²)(°F) for the natural convection heater. The measured heat transfer coefficient for the forced convection heater is compared to correlations. Hot air heating also provides improved cylinder heating safety by increasing the unloading rates while operating at a low cylinder temperature, providing a large margin of safety against a hydraulic cylinder rupture. Administrative and engineered safety features to prevent high cylinder pressures and UF₆ leaks are discussed.

NOMENCLATURE

A_o	=	External surface area, ft ²
A_i	=	Internal surface area, ft ²
C_p	=	Specific heat, Btu/lb °F
d	=	Jet orifice diameter, in.
$dT/d\theta$	=	Rate of change of UF ₆ temperature with time, °F/min
g	=	Acceleration of gravity, 32.2 ft/sec
h_o	=	External film heat transfer coefficient, Btu/(hr)(ft ²)(°F)
h_i	=	Internal film heat transfer coefficient, Btu/(hr)(ft ²)(°F)
k	=	Thermal conductivity, Btu/(hr)(ft ²)(°F)
L	=	Characteristic length, ft
M	=	UF ₆ mass, lb
m	=	Reynolds number exponent (1)
q	=	Rate of heat transfer, Btu/hr (cal/min)
U	=	Overall heat transfer coefficient, Btu/(hr)(ft ²)(°F)
S	=	Distance from nozzle opening to solid surface, in.
ΔT	=	Temperature difference between inlet air (forced convection heater) or heater wall (natural convection heater) and UF ₆ temperature, °F
Δt	=	Temperature difference between cylinder surface and fluid, °F

μ	=	Viscosity, lb/hr-ft
ρ	=	Density, lb/ft ³
β	=	Volumetric coefficient of thermal expansion, 1/°F
ϕ_1	=	Adjustment factor based on pitch pattern (1)
ϕ_2	=	Adjustment factor based on number of rows in direction of spent flow (1)

INTRODUCTION

Production of nuclear fuels at the Savannah River Plant (SRP) requires highly enriched uranium hexafluoride (UF₆) to be heated and vaporized. The objective of the UF₆ heating system is to safely unload the cylinder in an acceptable amount of time. Process upsets such as pluggages and UF₆ leaks must be prevented to prevent production downtime and ensure a safe operation. This goal has led to the development and installation of a recirculating forced air convection heater to improve the safety and operability.

EQUIPMENT DESCRIPTION

Natural Convection Heater

The original heater in use at SRP was constructed from a 24-in.-high, 6-in.-dia metal pipe with a flat bottom. The UF₆ cylinder was centered in the heater with a 0.4-in. gap between the outer cylinder wall and the inner heater wall. The heater shell and bottom were electrically heated to the desired operating temperature. A removable insulation jacket was placed around the cylinder top and valves to prevent heat loss and pluggages. The UF₆ cylinder was connected to the downstream piping manifold by means of a flexible, heat-traced copper pigtail. The cylinder surface, heater wall, and heater bottom temperatures were measured for control and overtemperature protection.

Forced Convection Heater

The forced convection heater system consists of a multichambered shell containing air supply and return plenums to recirculate hot air to the UF₆ cylinder (Fig. 1). Hot air is supplied by a blower and heating element at the rate of about 125 ft³/min. During heating, a hinged tophat is closed and sealed so that the entire cylinder and pigtail connections are enclosed in the heater. The heater is enclosed in a glovebox to protect personnel from potential UF₆ leaks. The entire heater and associated ductwork are insulated to minimize heat loss and to protect personnel from thermal burns.

The vertical air supply plenum is 2 ft in length and reaches a point just below the cylinder handles. The vertical plenum contains 1680, 1/8-in.-dia air supply holes spaced on a 1/2-in. square pitch. The cylinder is centered in the heater, creating a 3/8-in. air gap between the cylinder and heater surface. For every 24 supply holes there is a 3/8-in. exhaust hole on 2-1/2-in. centers connected to the return plenum. To

ensure that the cylinder top and valves are sufficiently heated, 120 supply holes are located in the tophat section in addition to a small tube that directs hot air directly on the cylinder unloading valve. The base of the heater contains an additional 76 supply holes. Air distribution plates are designed to give a relatively uniform air velocity through the holes. Multiple temperature sensors (RTD's) are located in the air distribution plenum and the inlet and outlet exhaust ducts for temperature control, high temperature interlocks, and process control.

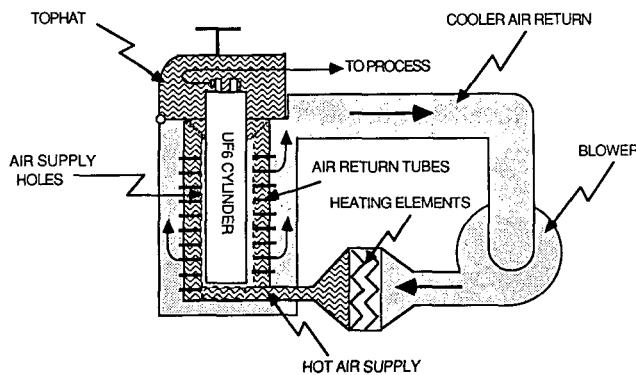


Figure 1. Schematic of Forced Convection Heater

CYLINDER UNLOADING OPERATION

To begin the unloading operation, a cylinder is placed in the center of the heater. The cylinder unloading valve (without diptube) is connected to the unloading manifold piping by means of a heat-traced copper pigtail. A clamp secures the unloading valve to the heater shell to prevent cylinder rotation and damage to the pigtail when the cylinder valve is opened. The manifold and pigtail connections are then pressure-checked with nitrogen to a minimum of 60 psig, and subsequently the lines are evacuated and a vacuum leak check performed. A minimum acceptable nitrogen leak rate of $0.025 \text{ cm}^3/\text{sec}$ is verified before cylinder heating is initiated. This ensures no inleakage of air during the time the cylinder is under a vacuum and no UF_6 leakage when the cylinder is under pressure.

The cylinder is then opened to the closed manifold. Since a cylinder of UF_6 at room temperature containing a minimum of nonvolatile compounds (air, nitrogen, HF) is under vacuum, a decrease in the manifold pressure should be observed when the cylinder valve is opened. This pressure drop is verified to ensure that the operator has actually opened the cylinder valve. If the pressure drop is not observed or the cylinder pressure is not less than 10 psig, the cylinder is evacuated (cold-burped) and the valve opening procedure repeated. The cylinder vacuum is later checked to ensure that no inleakage from a valve packing has occurred. An adequate vacuum prior to heating the cylinder is essential in ensuring that high cylinder pressures do not occur as a result of thermal expansion and compression of nonvolatile gases during heating. Compression of these gases occurs due to the large increase in UF_6 volume at the solid-liquid phase change in addition to the further increase in liquid UF_6 volume with temperature, reducing the free volume in the cylinder and manifold.

The cylinder is heated with the heater output controlled to maintain an air temperature of 100°C . The cylinder pressure is allowed to rise until a specified pressure, typically 45 psig, is reached. The manifold outlet valve is then opened, and the UF_6 is discharged to the process until the pressure decreases to 10 psig. At this point the manifold outlet valve is closed. The pressure then rises until 35 psig is reached, and the valve is

reopened. This procedure of heatup and unloading continues for 10 to 12 cycles until the cylinder is empty. The peak cylinder pressure and therefore UF_6 temperature is limited during each heatup cycle to maximize the difference in the air and UF_6 temperature, optimizing the heat transfer rate. The minimum cylinder pressure above the 7.4-psig triple point pressure prevents resolidification and a reduction in heat transfer rate.

ENGINEERED SAFETY FEATURES

Cylinder unloading is controlled and monitored by a Distributed Control System (DCS), which is interactive with the operators. In addition to standard process control functions, the DCS also displays process alarms, current trending of analog data, and provides safety interlocks. Independent hardwired interlocks are provided outside of the DCS for critical safety systems. In the forced convection heater, the air temperature is controlled by a temperature probe (RTD) located in the inlet air supply plenum. A DCS software interlock de-energizes the cylinder heater if the air temperature exceeds 115°C . Two additional RTD's are located in the inlet air supply plenum to provide independent hardwire interlock shutdown if the temperature exceeds 115°C .

A pressure transmitter, in addition to a manual pressure gage, located in the unloading manifold, monitors the pressure during cylinder heating. The heater is interlocked off if the pressure exceeds 80 psig during heating. The pressure transmitter also serves a second function to ensure an open cylinder valve during heating by de-energizing the heater if a minimum pressure rise is not detected during the first heatup cycle. To prevent a pluggage between the cylinder and manifold due to solidification of UF_6 vapor, the pigtail heat-tracing current is monitored, and an interlock will shutdown the heater if a drop in current is detected.

Emergency shutdown in the event of a UF_6 leakage is provided by a CO_2 cooling system. The system, which can be activated manually by the operator, delivers liquid CO_2 to two nozzles (located in the cylinder heater tophat) directed towards the cylinder unloading valve. Rapid cooling of the valve results in freezing of the UF_6 , preventing any further release. A rapid and slow CO_2 discharge from each nozzle respectively maintains the "freeze off" until additional shutdown measures can be taken. The glovebox also contains ionization-type "smoke" detectors to detect a UF_6 leakage. In addition to audible alarms to notify operating personnel, the CO_2 will automatically discharge if a failure of the ventilation scrubbing system occurs simultaneously with a detector alarm.

DETERMINATION OF OVERALL HEAT TRANSFER COEFFICIENTS

The instantaneous heat transfer rate is calculated by the rate of UF_6 temperature rise during heating:

$$q = M \cdot C_p \cdot dT/d\theta^* \quad (1)$$

The UF_6 temperature is calculated from the vapor pressure of UF_6 assuming liquid-vapor equilibrium. All experimental determinations are based on liquid phase UF_6 . Data from the first cylinder heatup prior to any discharge of UF_6 are not used. It is assumed that after the first unloading cycle, pure UF_6 remains and the cylinder pressure is equivalent to the vapor pressure. The slope of the cylinder pressure with time then

*Since a portion of the UF_6 cylinder is cooled due to the drop in UF_6 temperature during unloading, the heat transfer rate is actually higher than calculated from equation (1). The heat input required due to changes in the cylinder temperature has been neglected and is not included in the calculation of q in equations (1) or (2).

provides the instantaneous rate of temperature rise. The mass of UF_6 in the cylinder at any given time is calculated from the amount of UF_6 removed from the cylinder based on downstream processing conditions after each of the unloading cycles previously described. The heat transfer rate during each heatup cycle is then determined at a point when the UF_6 temperature reached 75°C (16.3 psig).

The overall heat transfer coefficient, U , is determined where the rate of heat transfer is

$$q = U A_0 \Delta T. \quad (2)$$

The area, A_0 , has been defined as the external cylinder surface area in contact with the liquid UF_6 , determined from the mass (volume) of UF_6 remaining in the cylinder. The external surface area and the instantaneous heat transfer rates were then determined during each heatup cycle. The slope of the heat transfer rate divided by the temperature difference versus the external surface area is then the desired overall heat transfer coefficient.

RESULTS AND DISCUSSION

Production data for the two heating systems were analyzed to determine the heat transfer rate for the natural convection versus forced convection heater. Cylinder pressure is shown as a function of heatup time for the two systems in Fig. 2 for the first two heatup and unloading cycles. A direct comparison can be made of the time required to liquify all of the UF_6 in the cylinder. The time when the UF_6 begins to melt at the triple point is evidenced by a decrease in the rate of pressure rise during the first heatup cycle. The triple point (64°C) is reached in 35 minutes with the forced convection versus 140 minutes with natural convection. The time when all of the UF_6 has melted is indicated by an increase in the rate of pressure rise, which occurred at 90 minutes for the forced convection and 350 minutes for natural convection. The total time required to melt the UF_6 is then 55 minutes and 210 minutes, respectively. The ratio of 3.8 is then an indication of the improvement in the heat transfer rate for the forced convection heater. This is only a rough estimate, and no determination of the overall heat transfer coefficient was attempted using these data.

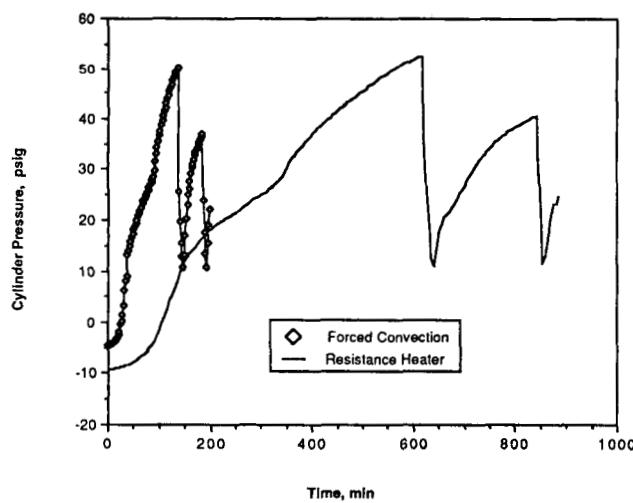


Figure 2. Rate of Pressure Rise during Heating

The pressure continues to increase during the time required to liquify the UF_6 indicating two phases and a nonuniform temperature distribution in the cylinder. The average UF_6 temperature and the cylinder surface area in contact with the UF_6 are difficult to predict with any accuracy. The porous nature of the solid UF_6 , difficulty in predicting the percentage of solid and liquid during fusion, and the large decrease in density at the phase change prevent an accurate determination of the overall heat transfer coefficient.

To provide a more accurate comparison and actual values for the overall heat transfer coefficient, the method described previously was employed. The data in Table 1 are a summary of heat transfer data during each heatup cycle at a point when the UF_6 temperature was calculated to be 75°C . The temperature difference is therefore 25°C between the cylinder contents and the heating medium. In Fig. 3, the heat transfer rate as a function of surface area is presented with the slope equal to the overall heat transfer coefficient. For the forced convection heater a value of U of $11.5 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F})$ was calculated versus $3.9 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F})$ with the original natural convection heater, for a factor of three improvement.

The increase in the heat transfer is explained by the different principal mechanisms for the external heat transfer coefficient for the two heating systems. In the natural convection heater, heat transfer from the heater to cylinder is across a small air gap. Natural convection occurs due to the difference in temperature between the heater and cylinder surface. In addition to natural convection, the contribution to the outside heat transfer coefficient from radiative heat transfer is expected to be on the same order of magnitude as that from natural convection.

In the recirculating hot air heater, the relatively high air velocity created by the small impingement holes changes the mechanism for heat transfer to forced convection. The convective heat transfer coefficient between a solid surface and an array of gas jets normal to it may be found from equation (3), Ref. (1). All properties are at the orifice outlet. The quantities m , ϕ_1 , and ϕ_2 are graphically presented:

$$h = \phi_1 \phi_2 12 k N_{\text{Re}}^m (S/d)^{0.091} N_{\text{Pr}}^{0.333/d}, \quad (3)$$

Table 1. Heat Transfer Rate during Each Heatup Cycle

Heatup Number	$dT/dt @ 75^\circ\text{C}$	UF_6 Mass	External Surface Area	Heat Transfer Rate, Q
	°F/min	lb	ft ²	Btu/hr
Natural Convection Heater				
1		53.2		
2	1.07	34.7	1.83	308
3	1.13	23.7	1.31	183
4	1.17	16.5	0.97	155
5	1.21	9.0	0.62	86
Forced Convection Heater				
1		53.6		
2	3.01	38.3	2.00	908
3	3.91	27.3	1.48	841
4	4.18	20.7	1.17	682
5	3.15	15.2	0.91	377
6	4.01	11.4	0.74	362
7	4.01	7.9	0.57	250
8	4.03	5.2	0.44	165

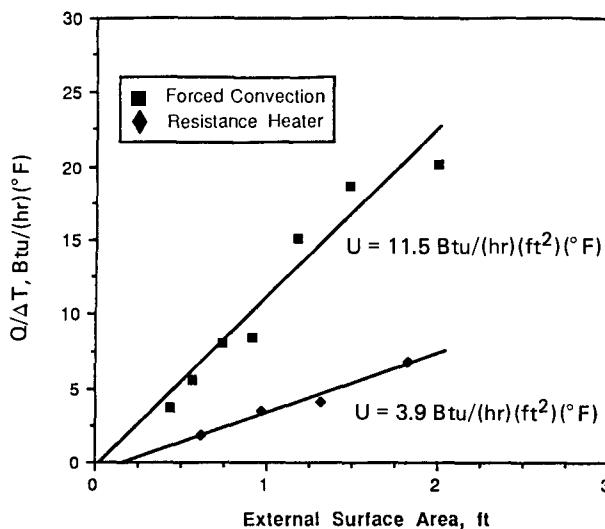


Figure 3. Comparison of Overall Heat Transfer Coefficient

where:

$$N_{Re} = \rho V d / 12 \mu.$$

$$N_{Pr} = C_p \mu / k.$$

The blower discharge rate was experimentally determined by measuring the rise in air temperature across the heating elements and the power supplied to the heater. For a heater output of 2500 W and a temperature increase of 32°C, this corresponds to 125 scfm. Air velocity through the impingement holes normal to the cylinder surface is about 16 ft/sec. The correction factor α_2 , which reduces the heat transfer coefficient due to interference of exhaust air, is assumed to be one because of the large number of evenly spaced exhaust holes. The calculated value for the external heat transfer coefficient is 22 Btu/(hr)(ft²)(°F). Using the experimentally determined value for U , the internal heat transfer coefficient is calculated to be 26 Btu/(hr)(ft²)(°F) where

$$1/U A_O = 1/h_O A_O + 1/h_i A_O . \quad (4)$$

An estimate of the internal heat transfer coefficient can be made assuming the well known correlation for natural convection for a vertical surface,

$$N_{Nu} = 0.13 (N_{Gr} N_{Pr})^{1/3},$$

where:

$$N_{Gr} = L^3 \rho g \beta \Delta t / \mu,$$

$$N_{Pr} = C_p \mu / k,$$

$$N_{Nu} = h L / k,$$

with physical properties evaluated at the film temperature.

With the average UF_6 temperature during a heatup cycle estimated to be 85°C, the calculated value for the internal heat transfer coefficient, h_i , is 100 Btu/(hr)(ft²)(°F). Using the calculated value of h_o , the overall heat transfer coefficient from equation (4) is then 18 Btu/(hr)(ft²)(°F), or a factor of 1.6 higher than the experimentally measured value. The experimentally determined overall heat transfer coefficient may be underestimated due to the effect of the exhaust air. The average between the air supply temperature and the exhaust temperature may be more appropriate to use in equation (2). This results in a ΔT of 21°C and a measured value for U of 13 Btu/(hr)(ft²)(°F), approximately 25% less than the calculated value.

ACKNOWLEDGMENT

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SAFETY AND SECURITY IMPROVEMENTS IN THE PGDP UF₆ SUBSAMPLING LABORATORY

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ABSTRACT

To provide additional assurance that nuclear materials are being handled safely and securely, the UF₆ Subsampling Laboratory at the Paducah Gaseous Diffusion Plant has made some major changes in operating methods. Three systems have been redesigned to improve safety, and a near real-time inventory system has been devised to maximize the security of nuclear materials while in the lab. To handle the return of excess UF₆ from samples back to the cascade, a new dumping system was designed and installed by plant personnel which eliminates the use of the open-flame torch as a heat source in the operation. The new system uses a combination of self-limiting heat tape and temperature controls with high temperature alarms. Similarly, the subsampling manifold used to take small aliquots for the various analytical measurements from the larger bulk sample cylinders has been replaced with an electrically heated, temperature-controlled, and alarmed system. Again, the open-flame torch was removed. Preparation of the cylinders for reuse involves fluorination to passivate the inner surfaces. The new manifold system designed for this procedure is automated to eliminate the need for manual valving of fluorine into the system. Finally, a program has been written and installed on the lab computer sample tracking system enabling lab personnel to know the location and status of all 2S sample cylinders. The near real-time inventory requires only a few minutes of operator time each day to update cylinder status changes and makes it possible to complete an inventory of all sample cylinders in less than forty-five minutes. These improvements, with existing procedures and policies, assure the safe and secure handling of nuclear materials by lab personnel at the Paducah Gaseous Diffusion Plant (PGDP).

Prepared by the Paducah Gaseous Diffusion Plant, Paducah, Kentucky 42001, operated by MARTIN MARIETTA ENERGY SYSTEMS, INCORPORATED, for the U.S. DEPARTMENT OF ENERGY, under Contract No. DE-AC05-84OR21400.

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INTRODUCTION

The PGDP UF₆ Subsampling Laboratory is responsible for: 1) subsampling UF₆ from 2S cylinders into smaller containers suitable for various laboratory analyses, 2) dumping the excess UF₆ from 2S cylinders into 12A cylinders for refeeding to the cascade, and 3) cleaning and fluorinating the 2S cylinders for reuse. Even though this type laboratory handles relatively small quantities of UF₆ as compared to cascade operations, there are many of the same safety concerns. To provide additional assurance that safety guidelines are being met, improvements have been made in each of the above mentioned functions. Due to the increased emphasis on security of small quantities of nuclear materials, a computer sample cylinder tracking system and additional physical controls have been added.

SUBSAMPLING

Subsamples for the various analytical measurements are taken from bulk sample containers (usually 2S cylinders). An open-flame torch had previously been used to supply the heat necessary to subsample UF₆. Because this type of torch is an uncontrolled heat source, an alternate method of heating was sought. Flameless subsampling was demonstrated by Hall, Hedge, and Reid through the use of controlled heating tape and a cartridge heater.(1) Using the same concepts, a system was configured by Subsampling personnel which fit the subsample tube and space requirements needed. Safety features of the system include:

1. Self-limiting heat tape and cartridge heaters
2. Manifold fabricated and inspected to UF₆ pigtail specifications
3. Temperature control
4. Audible and visible alarms
5. Automatic system shutdown
6. Redundancy in power control

DUMPING

The 2S cylinder samples which are no longer needed are dumped into 12A cylinders which can be refed

to the cascade. This system, like the subsampling system requires a heat source. The open-flame torch has been replaced with canister heaters and heat tape. Safety features include:

1. Self-limiting heat tape
2. Eleven monitored points with audible high temperature alarm
3. Dual ventilation system

FLUORINATION

After the 2S cylinders have been cleaned, they are passivated with fluorine before they are returned to service. An automated fluorination system has been installed which allows the laboratory analyst to perform this operation within an enclosed hood. Safety features include:

1. Automated operation
2. Pump-down interval alarms
3. Self-limiting heat tape
4. Remote operation of fluorine valves
5. Fluorine flow sensor and automatic shut-off

CYLINDER TRACKING SYSTEM

With increased emphasis on nuclear material physical security, an efficient means of inventorying 2S sample cylinders was needed. A computer tracking system which uses existing laboratory computer equipment was implemented for this purpose. The following steps summarize the program operation:

1. All 2S cylinders are entered in the program by their cylinder number.
2. The cylinder number is coded to give additional information about the cylinder status (e.g., material type, control, etc.).

3. The location or person responsible is entered.

4. A dated entry is made in one of four categories:

READY FOR USE
SENT TO C-360
IN USE
DUMPED

5. The program is updated daily, or as needed, as cylinder status changes.

6. A status listing compiles all of the information, including a container count.

This near real-time inventory allows an inventory of all 2S cylinders in the plant to be made and verified in 30 to 45 minutes. Daily input to the system usually takes less than five minutes--much shorter than other physical means of tracking the cylinders would be. Also, the program is written to be flexible so that additional information, such as hydrotest dates or other cylinder types, can be included.

SUMMARY

Recent changes in the Subsampling Laboratory operations provide additional assurance that the handling of UF_6 is performed in a safe manner. The use of a computer cylinder tracking system provides an expedient near real-time inventory which is flexible for future changes.

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USE OF TAMPER INDICATING DEVICES (TID) ON UF₆ CYLINDERS

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ABSTRACT

Tamper Indicating Devices (TID) have been used at the Paducah Gaseous Diffusion Plant (PGDP) for a number of years to indicate the authenticity and to verify the integrity of UF₆ cylinder shipments.

All shipments involving any nuclear material container, including shipments of empty cylinders, are sealed with an accountable TID. An unviolated TID permits verification of the contents of the container. Conversely, a violated seal is an indication of possible tampering.

Several alternative designs were developed and tested. A cost effective TID was designed and implemented that achieved high quality and met objectives of indication of tampering of the valve and valve part.

TID CHARACTERISTICS

- A. **Frangible** - Seals are easily broken and are not intended as deterrents to an adversary willing to use force but will identify if tampering has occurred.
- B. **Nonreversible** - Once a seal is broken, it is difficult to reassemble without leaving signs of tampering.
- C. **Identifiable** - Seal must be distinguished by unique identification characteristics, such as serial numbers to remain traceable throughout the system.

HISTORY OF THE USE OF TIDS AT PGDP

A number of different type TIDs have been used at the Paducah plant over the years. Lead nonnumbered seals on the valve protector was the first TID used (Figure 1). Plastic bags with the seals on the inside of the bag were tested for a

period of time (Figure 2). A reinforced bag with fiberglass strands woven in the bag was also tested. This bag could not be torn and would show signs of tampering if a wrench was used to unscrew the valve. During the testing of this bag, it was discovered that when the temperature was below ten degrees fahrenheit, the glued seam would pull apart (Figure 3).

Various other plastic bags were tested using wire seals (Figure 4). The main problem found with the plastic bags was the difficulty in threading the wire through the six prepunched holes and attaching the seal under adverse winter conditions.

During the development efforts for a new design, the following objectives were considered: high quality, low cost, virtually impossible to access any part of the valve body without detection, and ability to install the TID under adverse winter conditions with gloves on.

The first TID tested was made from a section of schedule 40 PVC pipe with a solvent welded PVC cap on one end. This TID version was discarded due to the high cost of the material and labor to fabricate the TID (Figure 5).

The TID now being used at PGDP is a one-piece, seamless, tapered polypropylene plastic cover (Figure 6) attached to the cylinder valve with a metal pin and numbered seal (Figure 7).

Nuclear Material Control and Accountability (NMC&A) must be combined with TID usage. All shipment and receipt documentation of UF₆ cylinders must contain the TID number.

All full and empty UF₆ cylinders being shipped are inspected to verify that the TID is properly applied and the TID number is on the nuclear materials tally-out sheets and bill of lading.

This prototype design is proposed for industry-wide use. The TIDs must be accepted by all and somewhat consistent in design for the TID system to work.

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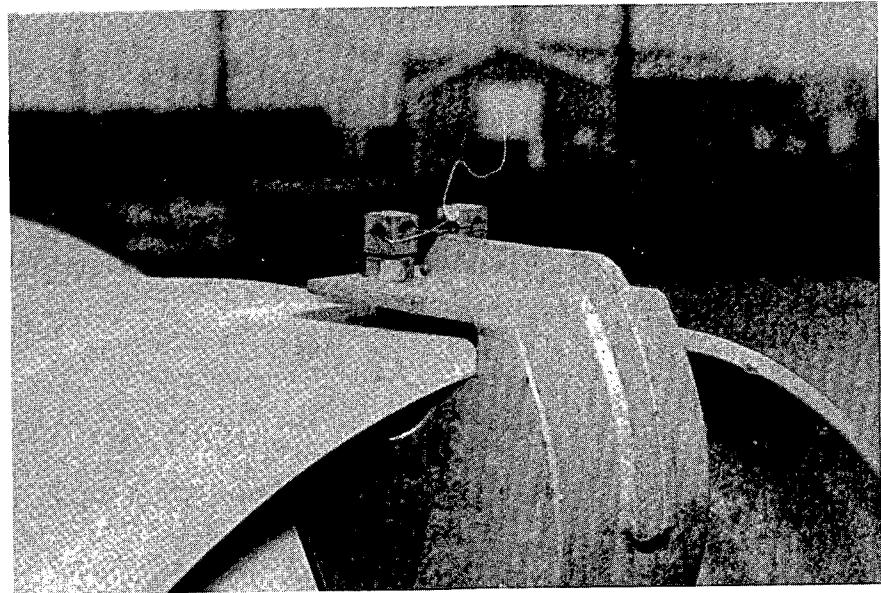


Figure 1. VALVE PROTECTOR WITH LEAD SEAL

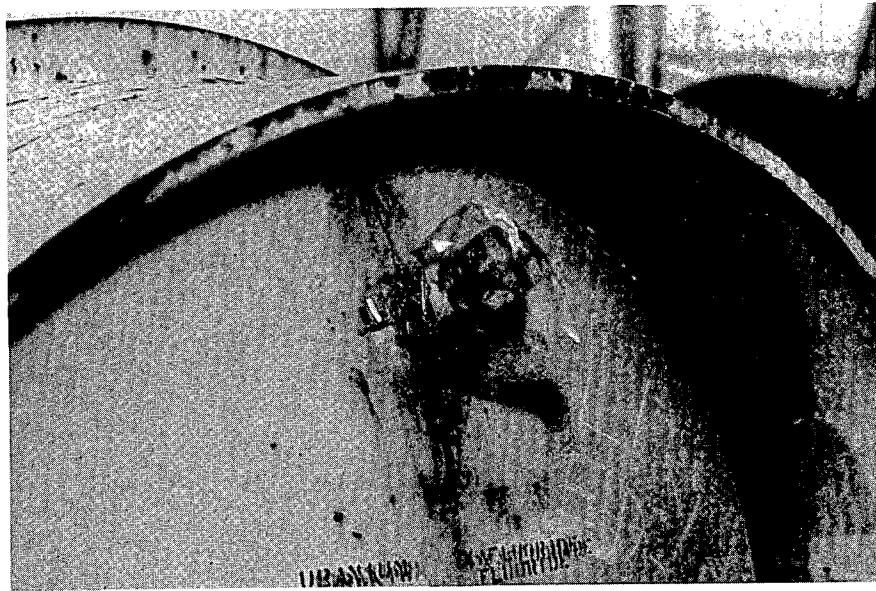


Figure 2. CLEAR PLASTIC BAG

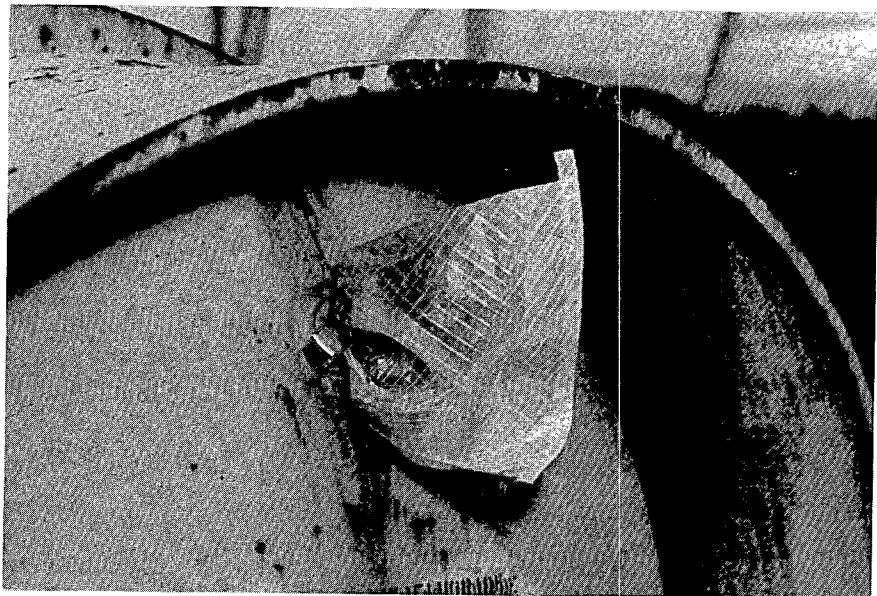


Figure 3. REINFORCED PLASTIC BAG

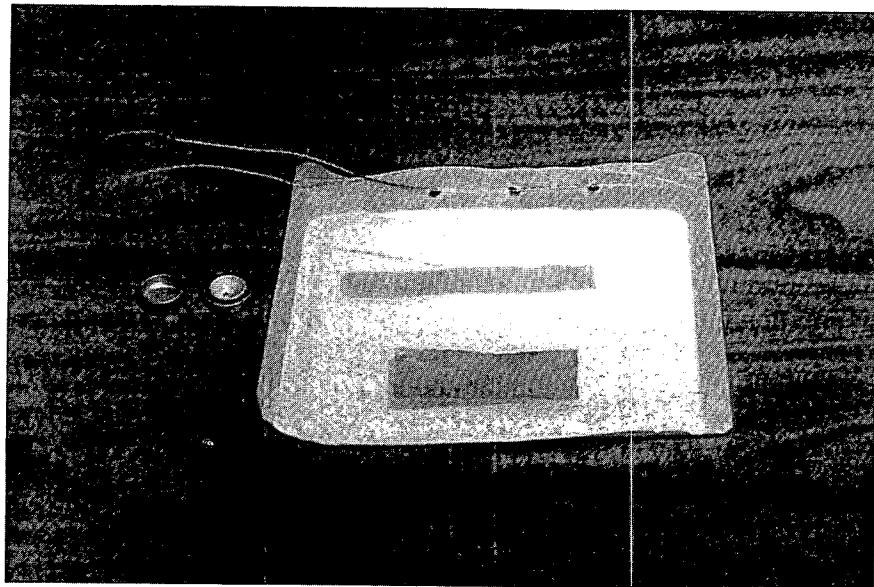


Figure 4. PLASTIC BAG WITH PREPUNCHED HOLES

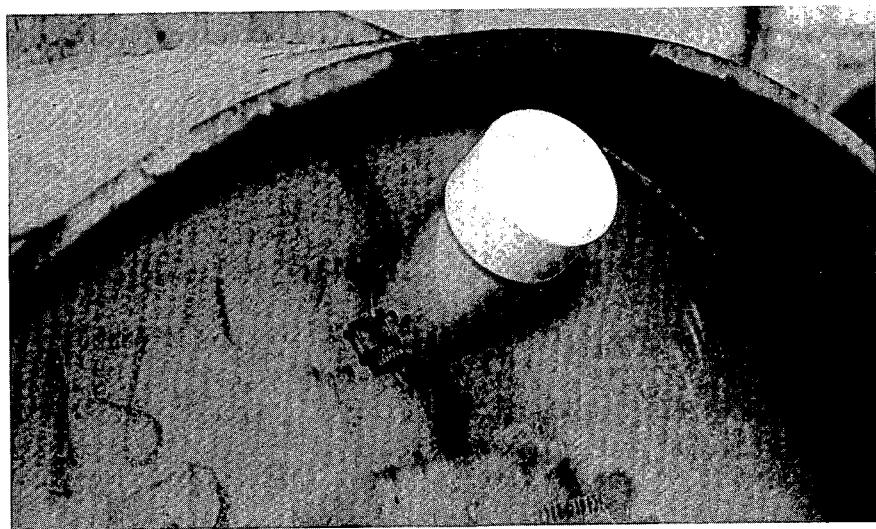


Figure 5. PVC PIPE AND CAP

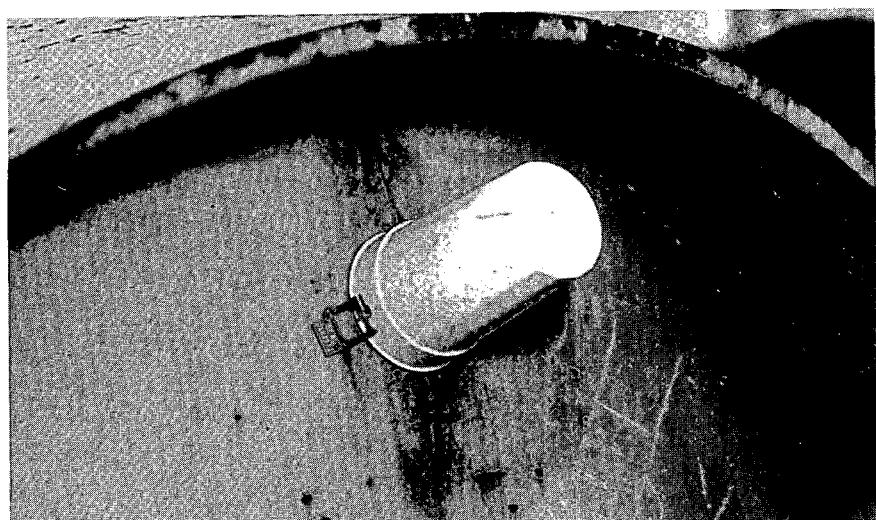


Figure 6. ONE PIECE SEAMLESS TAPERED PLASTIC COVER

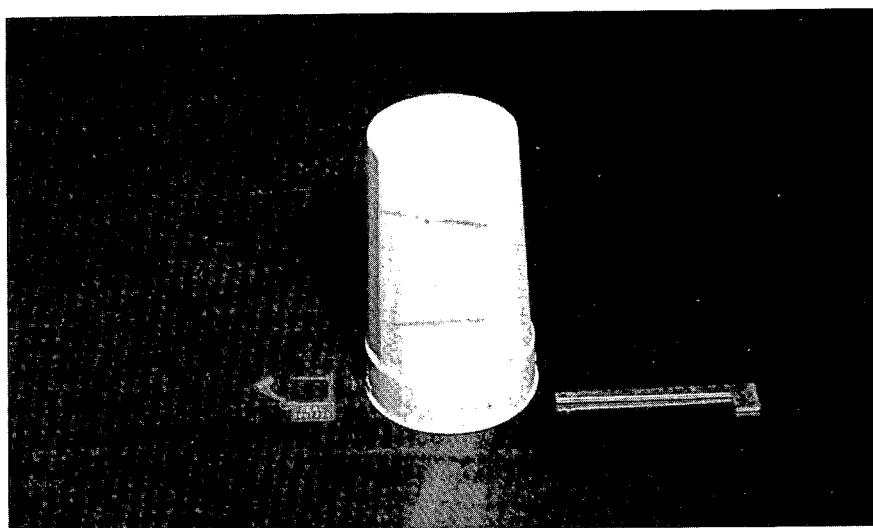


Figure 7. LOCKING PIN AND SEAL

DEVELOPMENT OF A 20-TON-CAPACITY LOAD-CELL-BASED WEIGHING SYSTEM FOR IAEA FIELD USE¹

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ABSTRACT

A 20-ton-capacity Load-Cell-Based Weighing System (LCBWS) is being developed to provide the International Atomic Energy Agency (IAEA) with a portable means of verifying the masses of 10- and 14-ton UF₆ cylinders at UF₆-bulk-handling facilities subject to IAEA safeguards inspections. The system is being designed to meet IAEA objectives for portability and accuracy as well as to meet facility operating safety conditions. During the development effort, system design requirements were established, and a comprehensive survey of commercially available weighing equipment against these requirements was conducted. Detailed equipment specifications for 20 load cells from 13 manufacturers, for 5 digital crane scales from 3 manufacturers, and for 7 digital weight indicators from 5 manufacturers were compared. The results of the survey favored a configuration based on a custom-designed load cell integrated with a commercially available digital weight indicator. Safety certification of the system by an internationally recognized verification authority is also recommended. Comparison criteria and their effect on the LCBWS design requirements are discussed, and the status of the project is reported.

INTRODUCTION

For the application of International Atomic Energy Agency (IAEA) safeguards at uranium enrichment and fuel fabrication plants subject to IAEA inspection, the IAEA requires an independent means of verifying the masses of UF₆ cylinders. Several portable 5-ton-capacity load-cell weighing systems have been developed with the capability of weighing a 2.5-ton UF₆ cylinder. These weighing systems have been in routine use by the IAEA. To provide the IAEA with a means of verifying the masses of 10- and 14-ton UF₆ cylinders at UF₆-bulk-handling facilities, a 20-ton-capacity Load-Cell-Based Weighing System (LCBWS) is being developed. The system is being designed to meet IAEA objectives for portability and accuracy as well as to meet facility operating safety requirements. Funding for this equipment development is being provided by the U.S. Program for Technical Assistance to IAEA Safeguards (POTAS) under Task A.137.

BACKGROUND

A prototype 20-ton-capacity LCBWS was developed jointly by Brookhaven National Laboratory and Oak Ridge Gaseous Diffusion Plant (ORGDP) (1). The prototype system was configured as two parallel branches, each of which was comprised of one 11.4-ton-capacity load cell, two flexures, and

associated connecting hardware. The electronics of the system consisted of two digital load-cell indicators, one standardizer, and two digital temperature indicators connected to resistance temperature detectors. The electronics of the LCBWS was integrated with a computer control system programmed (1) to guide an IAEA inspector through the LCBWS weighing procedure, (2) to prompt the electronic instruments for the data, (3) to correct all field readings to calibrated standards, and (4) to calculate the mass of a given UF₆ cylinder. In-house tests of the prototype system indicated overall accuracies of ± 3 kg or better.

The prototype 20-ton-capacity LCBWS was field-tested by the IAEA at the Eldorado Resources, Ltd., facility in Port Hope, Ontario, in September 1986 (2). The test involved repeated weighings of one 14-ton UF₆ cylinder with different LCBWS configurations (e.g., with and without flexures, with and without computer interface). System accuracy and measurement repeatability demonstrated by the LCBWS were excellent. Cylinder weights as determined by the LCBWS computer system were within 0.05% of the facility scale weight; cylinder weights from the direct indicator readings were within 0.10% of the facility scale weight. Measurement results were unaffected by the removal of the flexures from the load-cell branches. To make the LCBWS a more rugged and portable system for routine IAEA field use, several modifications to the prototype system were suggested by the IAEA participants including removal of the flexures, removal of the temperature indicators, replacement of the two load-cell indicators with a single summing unit, battery operation, and elimination of the computer control system.

SAFETY CONCERN

Another issue that was raised during the field test of the prototype LCBWS was the operational safety of the system. At the time of the field test, the IAEA was experiencing difficulties in using their 5-ton-capacity load-cell system in European facilities. The 5-ton system was certified for 1.5 times its rated load. The British requested that the IAEA test each individual component of the system to two times its rated load prior to its acceptability for use. The Germans requested a similar certification criteria.

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Upon investigation, it was discovered that no uniform set of international standards for structural safety factors exists that applies to mechanical lifting hardware. The applicable American National Standards Institute, Inc., standard [i.e., ANSI N14.6-1978, "American National Standard for Special Lifting Devices for Shipping Containers Weighing 10,000 Pounds (4500 kg) or More for Nuclear Materials"] requires that all components of structural appliances used to lift nuclear materials weighing 10,000 lb or more be designed to have a safety factor of 5 relative to the ultimate strength of the components. For purposes of sizing components, deadweight loads are the sum of the cylinder weight and any lifting hardware that will be acting on the weighing system. Deadweight loads should be increased by an additional factor to conservatively account for the inertia effects of raising the cylinders with cranes and forklifts or bump loads induced by transporting cylinders with the weighing system attached.

MODIFIED LCBWS DESIGN SPECIFICATIONS

Since their initial purchase, the load cells of the prototype 20-ton-capacity LCBWS have been downrated by the manufacturer from a safety factor of 5 to a safety factor of 3; this necessitated the replacement of the system load cells. Based on the system modifications identified during the IAEA field test of the prototype LCBWS and the inclusion of the ANSI N14.6 safety requirements, new design requirements for a modified 20-ton-capacity LCBWS were prepared. The new configuration will be designed to meet IAEA technical objectives for accuracy and portability as well as to meet facility operating safety conditions. In addition, the system is to be configured with as many commercially available components as possible to minimize development time and field maintenance. The expected accuracy achievable with the system is better than $\pm 0.1\%$ of the weight being measured. The critical components of the system will be transportable by one person. To provide flexibility in interfacing with facility lifting equipment, the weighing system will have the capability for single- or double-branch configuration. The system will meet or exceed the safety standards at any UF_6 -bulk-handling facility where its use is anticipated. Safety certification of the modified system by an internationally recognized verification authority is recommended.

EQUIPMENT SURVEY

Upon recommendation from the International Safeguards Project Office (ISPO), a comprehensive survey of commercially available load cells and digital weight indicators for the 20-ton-capacity LCBWS was conducted. Five commercially available crane scales were also included for comparison. Detailed equipment specifications for 25 load cells from 15 manufacturers, for 5 digital crane scales from 3 manufacturers, and for 7 digital weight indicators from 5 manufacturers were compared. Comparison criteria for the weighing equipment included ultimate load factor, safe overload factor, rated output, accuracy, load cell weight, configuration, cost, and availability. Comparison criteria for the digital weight indicators included interchangeability, count resolution, integral standardizer feature, weight, cost, and availability.

WEIGHING EQUIPMENT COMPARISON

Table 1 summarizes the specification ranges for the 5 crane scales and 25 load cells compared in the survey. The load cells are grouped into three different configuration categories based on shape: barrel, flat disc, and tension link (Figure 1). The barrel-type load cell is the most common type representing 15 of the 25 cells surveyed. Disc-type and tension link load cells are evenly represented at five cells each.

In selecting load cells and crane scales for comparison, a maximum lifting load of 20 tons was assumed. By sizing an individual load cell to accommodate the largest lifting load anticipated for the LCBWS, the weighing system can be configured as a single-branch or double-branch system. An additional assumption was a system ultimate load factor of 7.5. (The system ultimate load factor is defined as the ratio of the weighing device's structural failure load to 40,000 lb.) The 7.5 ultimate load factor is based on the ANSI N14.6-1978 standard with a dynamic load factor of 1.5 to account for inertia effects. (Because safety factor requirements at foreign nuclear-material-handling facilities are unknown, the largest dynamic load factor requirement found in the industry, that of the Portsmouth Gaseous Diffusion Plant, was chosen as a conservative figure.) When the static and dynamic loads are combined, an overall system ultimate load factor of 7.5 results. The rated capacity of the commercial load cells and crane scales being compared ranged from 50,000 to 200,000 lb because of the different ultimate load factors used by the various manufacturers in designing their weighing devices. (Ultimate load factor is defined as the maximum load as a factor of rated capacity that can be applied without producing a structural failure.) The standard manufacturer's ultimate load factor for crane scales is 5; load-cell ultimate load factors ranged from 2 to 5. By multiplying the manufacturer's ultimate load factor of a device by its rated capacity and then dividing by the maximum system lifting load of 40,000 lb, a system ultimate load factor could be calculated.

As an indication of equipment ruggedness, safe overload factors were compared. Ranging from 1.5 to 2.0 for the crane scales and from 1.5 to 3.0 for the load cells, the manufacturer's safe overload factor is defined as the maximum load as a factor of rated capacity that can be applied without causing permanent damage to the weighing device.

The larger the electrical signal produced by a load cell, the more sensitive the cell will be for small weight differences. The algebraic difference between the electrical signal produced by the load cell at its rated capacity and at no load, expressed in terms of mV/V , is known as the manufacturer's rated output. To meet safety specifications, many of the load cells being compared are oversized. Thus the signal output for these oversized load cells in the expected operating range of the LCBWS (i.e., 4500 to 34,000 lb, corresponding to the nominal tare weight of a 10-ton cylinder and the nominal gross weight of a full 14-ton cylinder) is substantially less than the cell's rated output. To compare the sensitivity of the various load cells, signal outputs at 40,000 lb were calculated.

Table 1. Crane scale and load-cell specifications

Crane scales (5)	Load cells			
	Barrel (15)	Disk (5)	Tension (5)	
Manufacturer's rated capacity (pounds)	50,000-77,000	50,000-200,000	100,000-125,000	80,000-100,000
Manufacturer's ultimate load factor	5.0	2.25-5.0	2.0-2.5	3.0-4.0
LCBWS ultimate load factor ^a	6-10	6.3-12.5	6.3-7.8	6.0-8.0
Manufacturer's safe overload factor	1.5-2.0	1.5-3.0	1.5	1.5-2.0
Manufacturer's rated output (mV/V)	-	2-6	2-4	2
Signal output at 20 tons (mV/V)	-	0.6-2.4	0.7-1.6	0.8-1.0
Accuracy	$\pm 0.1\%$ ± 10 lb to $\pm 0.1\%$ ± 44 lb	$\pm 0.37\%$ to $\pm 0.51\%$	$\pm 0.047\%$ to $\pm 0.24\%$	$\pm 0.15\%$ to $\pm 0.51\%$
Weight (pounds)	90-300	10-200	5-93	14-175
Length (inches)	20-43	7-18	2-10	10-19
Cost	\$6200-\$7000	\$795-\$5669	\$1150-\$5730	\$1030-\$2720
Availability	4-8 weeks	3-18 weeks	In stock - 8 weeks	4-8 weeks

^a Assumes maximum load of 40,000 lb.

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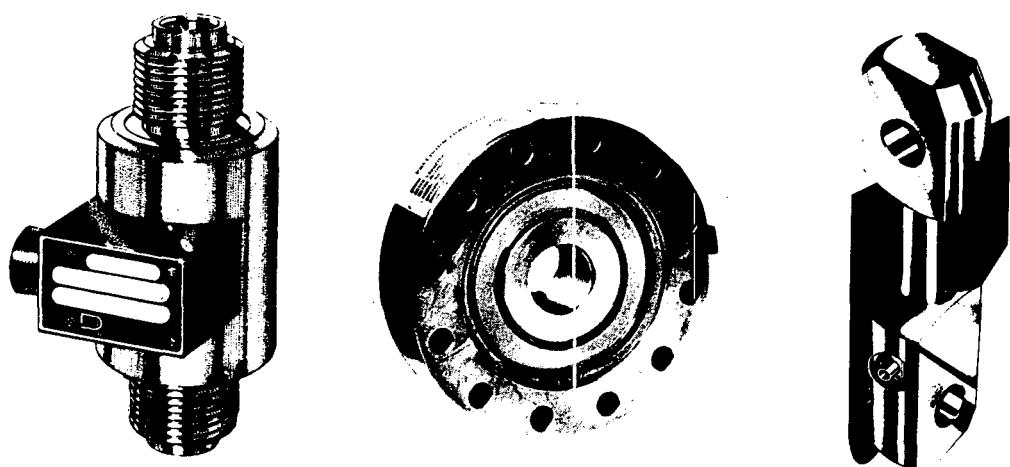


Figure 1. Load-cell configurations. (a) Barrel, (b) flat disc, and (c) tension link.

The accuracy of a load cell is defined in terms of nonlinearity, hysteresis, and repeatability. The load-cell accuracy ratings listed in Table 1 are based on the root mean square sum of these performance factors and are intended for use as a relative comparison figure only. The accuracy ratings for the crane scales are actual equipment specifications.

The weight and length listings in Table 1 refer to pounds and inches of the minimum lifting device configuration that could be transported. Crane scales ranged from 90 to 300 lb and from 20 to 40 in. Nondetachable swivel hooks and eye nuts on several models account for the figures in the upper end of each range. Weight varies substantially for each type of load cell. Both the barrel-type and disc-type load cells require separate attachments such as eye bolts, hooks, or shackles to interface them with a lifting device such as a crane or forklift (Figure 2). These attachments add approximately 60 lb to the reference weight and 10 in. to the reference length of both types of cells. The tension link load cells are configured with holes in their end plates for direct interface with lifting equipment.

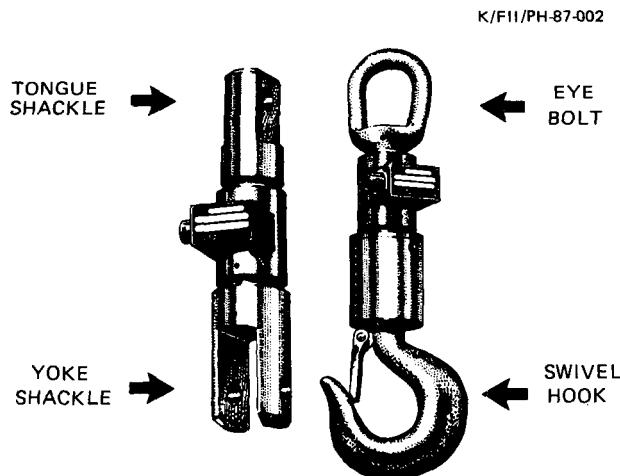


Figure 2. Sample load-cell interface attachments.

The cost entries in the table are based on the manufacturer's suggested retail cost for a single unit. The prices listed for the crane scales include the end hooks. The reference costs for the load cells do not include any attachments. A reference price for a pair of tongue shackles for the barrel- and disc-type load cells is approximately \$1000.

Availability refers to the time from receipt of order required for delivery. Because of the large rated capacity of the weighing devices being considered, the majority of the vendors do not have the devices in stock ready for delivery. In speaking with representatives of the various load-cell manufacturers while checking availability, many vendors offered to custom-design a load cell to meet the 20-ton-capacity LCBWS design requirements specifically. The cost and delivery time for a custom load cell was not substantially higher or longer than catalog-listed load cells.

INDICATOR EQUIPMENT COMPARISON

Seven digital weight indicators were compared for use in the LCBWS. All seven instruments were designed specifically for use in conjunction with load cells in weighing applications; therefore, all the instruments considered have common features such as push-button zero and motion detection. Table 2 summarizes additional design features for these weight indicators. The instruments are grouped into two different classes: Type I consists of microprocessor-based devices capable of in-field calibration; Type II involves simpler digital readout devices requiring laboratory calibration and matching of load cells and cables with the indicator. Two of the seven instruments fit into the first category; hence, design specifications are listed for both devices. Instrument capabilities considered important for LCBWS application are interchangeability, accuracy, and an integral standardizer feature.

The interchangeability feature implies that the instrument can be easily recalibrated in the field for any load-cell and cable combination. Constant, multiplicative correction factors for local gravity and buoyancy can be easily input from the front panel. Without this capability, end-to-end calibration of the load cell, the cable, and the weight indicator are required.

The accuracy achievable with a given instrument can be quantitatively estimated by the specifications of the instrument's voltage/frequency converter. For the indicators surveyed, two distinct classes of instruments were apparent (i.e., <20,000 count and 200,000 count). This specification can be interpreted as an expected accuracy of ± 4.0 lb for the 20,000-count device and ± 0.4 lb for the 200,000-count instrument. Clearly, if the ± 0.4 -lb-class instrument is used, instrument accuracy can be neglected when considering the overall LCBWS accuracy. The cost associated with this better resolution is nominally two to four times the cost of the lower accuracy instrument.

Table 2. Digital weight indicator specifications

Type	Programmable front panel	Count resolution	Integral standardizer	Battery option	Weight (pounds)	Cost	Availability
Type I (2)	Yes	200,000	Yes	No	8 and 20	\$3800 and \$5400	In stock
Type II (5)	No	10,000—20,000	No	No (4) Yes (1)	4—10	\$850—\$2000	In stock

The integral standardizer feature provides an in-field check of instrument accuracy by simulating the output of a load cell through a precision resistor network and provides a feedback into the instrument microprocessor for automatic meter drift compensation in the readout. Load-cell systems using a weight indicator without this feature require a separate standardizer in addition to manual interpolation of data.

Only one of the seven weight indicators has a manufacturer's supplied battery power option. The 8-lb microprocessor-based indicator is comparable in weight to the Type II digital readouts. The 20-lb weight indicator has an integral printer feature. All of the instruments surveyed are stocked by the vendors for immediate delivery.

RECOMMENDATIONS

Based on the equipment surveyed for the LCBWS, three different system configurations are possible: (1) a production crane scale, (2) a production load cell in conjunction with a production weight indicator, and (3) a custom load cell in conjunction with a production weight indicator. Option 3 was recommended as the base configuration for the 20-ton-capacity LCBWS. While a weighing system based on a commercially available digital crane scale has the advantages of being self-contained as well as battery operated, the system is very heavy (i.e., >75 lb) and has a limited accuracy capability (i.e., no better than ± 44 lb). Having no interchangeable parts, a crane scale is sensitive to singlepoint failure in the field. A weighing system based on a load cell in conjunction with a weight indicator has the advantages of being very portable, accurate, and versatile. Because of the low weights of the individual components (i.e., load cell <20 lb, indicator <10 lb), the system is easily transportable. Field maintenance is feasible with spare parts. Cost of the system is comparable to a crane scale; however, because of the capability for component replacement, maintenance costs should be less. Readily achievable system accuracies of better than $\pm 0.1\%$ will meet the current IAEA objectives for attributes measurements as well as future accuracy goals for variable measurements.

The recommended equipment for the modified 20-ton-capacity LCBWS includes a custom-designed load cell in conjunction with a digital weight indicator having an integral standardizer. Although there are catalog-listed load cells that meet the design requirements, a custom load cell manufactured to optimized design specifications can be lighter and more accurate. Cost and availability for a custom-designed load cell do not differ substantially from those quoted for the production units. The digital weight indicator with an integral standardizer permits the indicator to be used with any load cell for which calibration data is available; end-to-end calibration of the system is not required. Certification of the 20-ton-capacity LCBWS in conformity with various national safety standards by a technical surveillance authority is also recommended.

PROJECT STATUS

The load-cell equipment survey was reviewed with both ISPO and the IAEA in February 1987. Both concurred with the recommendation for a custom-designed load cell; however, a battery-powered weight indicator was preferred over the recommended weight indicator that featured an integral standardizer. Requests for bids on a custom-designed cell were sent to 14 load-cell vendors. The design specifications for the load cell included (1) a 40,000-lb capacity, (2) a weight of less than 40 lb, (3) a 2- to 3-mV/V output at 40,000 lb, (4) a safe overload factor of at least 1.5, (5) an ultimate overload capacity of 7.5, (6) an accuracy of better than 0.1%, and (7) a capability of meeting the ANSI N14.6-1978 standard. Following receipt and review of the vendor responses, the plan was to place an order for two identical cells and two battery-powered digital weight indicators. Assembly of the 20-ton-capacity LCBWS and in-house field tests of the system were then scheduled.

In March 1987 the IAEA decided to assemble a new 5-ton-capacity load-cell system instead of trying to upgrade its current system by use of a heavy safety yoke. An order was placed for a large quantity of German-made 5-ton-capacity tension link load cells and battery-powered digital weight indicators. In addition, three 20-ton-capacity tension link load-cell systems were ordered from the same manufacturer. The IAEA then arranged for a German surveillance authority to work with the load-cell manufacturer to certify the 5- and 20-ton load-cell systems in conformity with various national safety standards. In April 1987 ISPO notified Martin Marietta Energy Systems, Inc., that POTAS Task A.137 was officially on hold status pending a response from the IAEA on the status and operation of its new 20-ton-capacity load-cell systems. In December 1987 the material to be used by the manufacturer for fabricating the load cells was rejected by the certification authority after it failed an impact test. In January 1988 the new material passed all the required tests, and fabrication began. The current projection for delivery of the certified and calibrated German load-cell systems to the IAEA is April 1988. An offer has been made to the IAEA to field-test the 20-ton-capacity LCBWS at the ORGDP.

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FRACTURE CONTROL OF STEEL UF₆ CYLINDERS

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ABSTRACT

The deployment of the gaseous diffusion technology created a need for a large number of cost-effective and reliable vessels to store and transport uranium hexafluoride (UF₆). The diffusion plants have conducted destructive tests of the steel cylinders currently used in the UF₆ industry. Drop tests of the 10- and 14-ton cylinders, conducted at ambient and subzero temperatures, have demonstrated that they comply with the Code of Federal Regulation (CFR) governing the transportation of radioactive materials described in 10 CFR and 49 CFR. The fracture resistance of the cylinders was affected by structure features such as the thickness of stiffening rings and the presence of drain holes in attached skirts. Instances of cylinder failures are rare and have only occurred when a vessel was accidentally dropped or was involved in a collision with handling equipment. An investigation of a damaged cylinder revealed that the shells of some recently manufactured vessels could fail by lamellar tearing if they are impacted near a stiffening ring. The investigation was expanded into a study of the directional properties of steel used in new and old cylinders. On the basis of laboratory and model tests, the specification for cylinder steel was revised to improve its upper shelf impact resistance and reduce the potential for lamellar tearing.

INTRODUCTION

Today, there are thousands of steel cylinders owned by and in use at United States Department of Energy (DOE) facilities for the transportation and storage of UF₆. There are also thousands of cylinders privately owned by power companies, uranium producers, and other members of the international nuclear power industry. For decades the world-wide nuclear community has adopted the

designs used by the American gaseous diffusion plants. The immense quantity of material that had to be shipped and stored required designs that can be easily and economically fabricated from steel.

In the early days of the enrichment industry, the handling and transport of UF₆ was done with valves and cylinders designed for chlorine service. However, as the quantity of material rapidly increased it became necessary to develop a series of larger cylinders for transporting and storing UF₆. Since 1951, the enrichment plants have purchased approximately 50,000 10- and 14-ton capacity cylinders.

The following report will review the test work that has been done to determine the structural integrity of these cylinders, an investigation of a service failure, and a recent study that has led to the adoption of a tougher steel for the cylinder shell.

UF₆ CYLINDER TESTING

When a decision was made to transport UF₆ in thin-wall 10-ton cylinders on a one time basis, it became necessary to carry out destructive tests to ensure that their fracture resistance met the requirements of the Department of Transportation (DOT). Between 1965 and 1979, a series of cylinder drop tests were conducted at the Paducah plant.⁽¹⁻⁵⁾ Ten- and 14-ton cylinders were dropped onto small pistons and flat surfaces. Examples of the drop test activity and incurred test damage are shown in Figures 1 and 2. The following conclusions were drawn from the results of these tests.

1. The cylinders can sustain short drops onto a flat surface when the stiffening rings are deformed into the shell.
2. The unreinforced cylinder heads will withstand long drops onto a piston.
3. With the exception of the 10-ton thin-wall design, the shells experienced tears when they were impacted next to a stiffening ring.
4. The only instance of the formation of branching and running crack occurred when a cold, -13° F, 14-ton type 48HX thin-wall

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cylinder was dropped at a 35° angle from a height of four feet.

5. Only cylinders made from A285 steel were used in the drop tests.

These tests were designed and conducted to meet the Nuclear Regulatory Commission/Department of Transportation (NRC/DOE) license requirements for transportation of the cylinders. They did not address the conditions that could be encountered during in-plant cylinder handling or the threshold or limiting amount of impact damage that can be safely sustained.

CYLINDER FAILURES

The test programs established that 10- and 14-ton cylinders fabricated from A285 steel would withstand drops against flat surfaces. In March 1978 and March 1982, two failures occurred that involved the use of thin-wall cylinders made from A516 steel.

The steel change was made on the basis that it would improve the cylinder's fracture resistance if it was impacted at a temperature below the ductile brittle transition temperature of A285. After this change had been made, the shell of a 14-ton thin-wall cylinder was torn when it was accidentally dropped onto a wooden saddle. The investigation of the failure indicated that a sharp profile in the stiffening ring fillet weld could have intensified the impact force in the cylinder shell. Approximately four years later, a crack was discovered in the shell of another thin-wall cylinder made from A516 steel.

Investigation of the second failure led to the conclusion that the shells of cylinders made from A516 steel were subject to lamellar tearing if they were impacted close to the toe of the stiffening ring fillet weld (Figure 3).

Photoelastic analysis of two dimensional models of the stiffening weld joint revealed that there were high-stress concentrations through the thickness of the shell plate if the toe of the fillet weld was profiled to a sharp notch (Figure 4). The fillet profile observed on the second failed cylinder was also more convex than is normally found on typical UF₆ cylinders and contributed to the failure. When the fillet profile is concave, the highest stress is located away from the toe of the weld (Figure 5). The through thickness impact strength of the steel was less than 10 ft-lbs, and fracture faces of the impact specimens contained numerous, elongated sulfide inclusions characteristic of steel susceptible to lamellar tearing.

IMPACT TOUGHNESS OF CYLINDER STEEL

The specification for the fabrication of UF₆ cylinders has required that the steel used for the heads and shells shall have a minimum level of notch toughness at temperatures well below 0° F. This low temperature impact strength is a measure of the steels resistance to the initiation and propagation of brittle cracks. Specific levels of impact strength have been established by the testing of material from failed structures.⁽⁷⁾ For the lower strength carbon steels, the critical

levels of impact strength (Charpy V-Notch) are in the 10 to 20 ft-lb range. Currently, the specification for the A516 steel used for cylinders specifies that the material shall meet the requirements of ASTM A20. This ASTM specification requires that the minimum acceptable Charpy V-Notch energy absorption is 10 ft-lbs at a test temperature of -60° F. Cylinders made from material meeting this requirement would be resistant to brittle fracture in the event of an accident at very low temperatures. Unfortunately, the reliance upon the A20 requirement does not ensure that the steel will have the maximum possible impact strength to resist fracture by ductile tearing during the period when the cylinders are filled with liquid UF₆ or when the cylinders are being moved or loaded for storage or shipment. As shown in Figure 6, the decision to change from A285 Grade C to A516 Grade 60 has improved the low temperature impact strength of the cylinder steel. However, at the melting point of UF₆, the upper shelf impact strength of A516 is lower than that of A285. Also, the conventional steel-making practice used to produce A516 has reduced the through-thickness impact strength making the cylinders more susceptible to lamellar tearing if they are impacted near a stiffening ring.

A program was initiated in 1984 to determine if the use of low sulfur (<0.010%) grades of A516 would improve the impact resistance of the cylinder shell near the attachment of the stiffening ring. The low-sulfur steels, with inclusion shape control, are being used in many applications where there is a need for high notch toughness and resistance to lamellar tearing. An applicable example is found in a recent article on the use of low sulfur A516 for the fabrication of containers for the handling of nuclear material.⁽⁸⁾

A model was developed that simulated the geometric effects of the stiffening ring and cylinder saddle on the fracture resistance of the cylinder shell. The use of the model instead of a full size cylinder permitted the experimentation with material and design variables at a minimum of delay and cost. The test specimens consisted of four foot square plates, 5/16-inch thick, reinforced in the center with a thick, 10-inch diameter disc. The chemistry and mechanical properties of the plates are given in Table 1.

The toe of the fillet weld that attaches the disc was either blended into the plate to minimize any through thickness stresses or was notched with a disc grinder. The plates were fastened to a heavy steel frame which contained an inner ring that simulated the effect of the cylinder saddle (Figure 7). With the attached disc underneath, the back side was impacted with a 1,000-lb free-falling weight (Figure 8). The tests were conducted by a "staircase" loading of the plates. When a plate did not fail, it was struck again from a higher height. When a plate failed, the next one was impacted from a lower height (Figure 9). The fracture resistance of the plates is shown in Figure 10.

The drop height required to fracture the plates is a measure of the fracture initiation resistance of the stiffened weldment. The notching of the

fillet toe tended to reduce the fracture resistance of the high sulfur A516 and A285 plates. Smoothing of the fillet profile increased the height required to initiate failure, but the increased energy from the higher drops appeared to increase the amount of crack damage. The low sulfur plate of A516 was not only more resistant to crack initiation (higher drop height), but the higher toughness of the steel limited the size of the crack opening and the length of the crack. The low-sulfur steel failed by ductile tearing (Figure 11) while the conventionally melted A516 failed by delamination (Figure 12). The failure of the A285 plates to exhibit more impact resistance was probably due to its low through thickness impact strength.

Table 1

CHEMICAL COMPOSITION AND MECHANICAL PROPERTIES OF STEELS

Type	%C	%Mn	%P	%S	%Si
A285-GrC	0.21	0.50	0.014	0.019	-
A516-70	0.24	1.06	0.014	0.023	0.24
A516-70Ca	0.19	1.00	0.013	0.002	0.20
<i>C_u, ft-lbs, (3/4 size) 70°F</i>					
UTS (ksi)	YS (ksi)	Elong. (%, 8 in.)	Trans.	Short Trans.	
A285-GrC	66	46	25	44	10
A516-70	79	56	20	17	6
A516-70Ca	76	52	25	120	100

The adoption of a low-sulfur grade of A516, with inclusion shape control, would improve the impact resistance of UF₆ cylinders. It would not ensure the integrity of a cylinder dropped from several feet onto a saddle, but would offer more protection from damage due to drops under one foot and accidental impact from handling equipment and other cylinders. In the past, variation in the impact properties of conventionally melted steel made it difficult to forecast the fracture resistance of a cylinder type from the results of a single drop test. For instance, a 10-ton thin-wall cylinder appeared to have excellent puncture resistance; however, recent tests of steel from that cylinder have revealed that it had unusually high impact properties. The possibility of adopting a low-sulfur grade of A516 for cylinders was reviewed with several potential suppliers.

Three steel companies indicated that they could supply the steel at a cost of about three cents a pound above the current price of conventionally melted A516. The use of the improved steel for fabrication of the cylinder shell would increase the price of a cylinder by about fifty dollars, which is approximately 3%.

MODIFICATION OF CYLINDER STEEL SPECIFICATION

The specification for the thin-wall tails cylinder was changed to require the use of low sulfur, with inclusion shape control, for construction of the shell. A restriction was also placed on the profile of the fillet welds which attach the stiffening rings.⁽⁹⁾ The thinness of the shell (5/16 inch) makes it impossible to directly test the through-thickness properties without having to resort to the expensive methods of welding attachments to the upper and lower surfaces of the plates. At the suggestion of one of the mills which supply low-sulfur steel, the toughness of the shell was specified on the basis of the upper shelf energy for Charpy specimens oriented transverse to the rolling direction. Examination of published data and tests of cylinder steels indicated that the through-thickness impact strength increased rapidly when the transverse impact strength exceeded 50 ft-lbs (Figure 13). The cylinder specification was revised to require that the sulfur content not exceed 0.010% and that the transverse upper shelf impact strength not be less than 55 ft-lbs.

The impact strength of the steel produced from the first 176 cylinders of the 1985 order is shown in the lower, right-hand side of Figure 13. The impact strength of all of the plates exceeded the specified minimum.

CONCLUSIONS AND RECOMMENDATIONS

Studies of tests and failures of the steel UF₆ cylinders have been studied to determine if their fracture resistance can be improved with a minimum of impact on their design and costs. The following modifications were incorporated into the 1985 order for thin-wall cylinders.

1. The stiffening ring fillet welds on failed cylinders had convex profiles which formed a notch with the shell. The new specifications require that the fillet have a concave profile and blend into the shell.
2. The specification for the cylinder shell has been changed to require the use of low-sulfur steel made with inclusion shape control. This has increased the toughness of the steel.

It is recommended that the modifications to the thin-wall cylinders be included in the specification of the heavy wall shipping cylinders.

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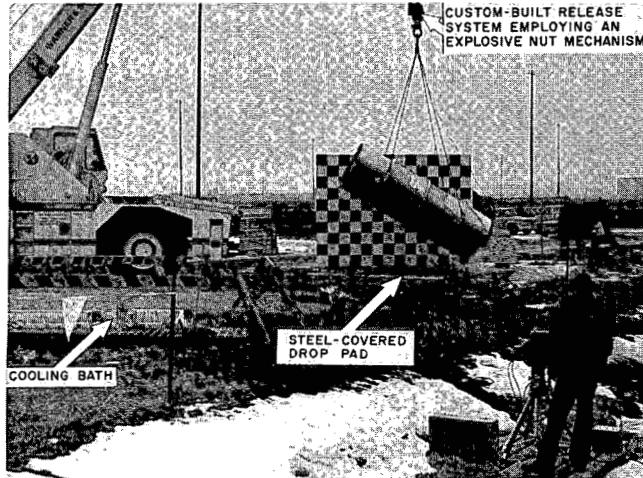


Fig. 1. View of Test Site with Cylinder
in Position for Drop

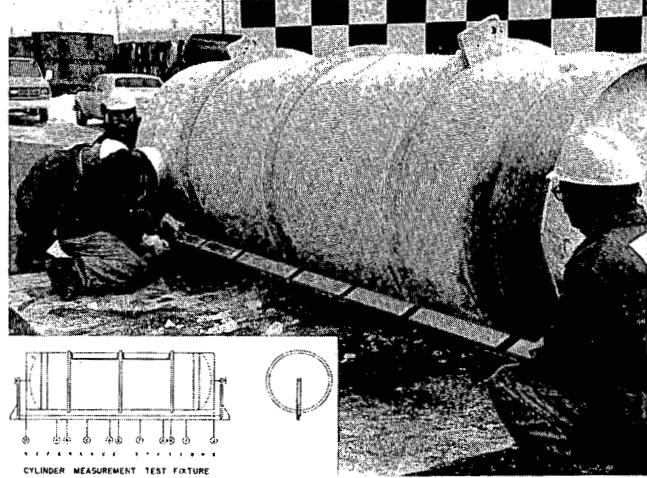


Fig. 2. Measurement of Cylinder Deformation
After Drop

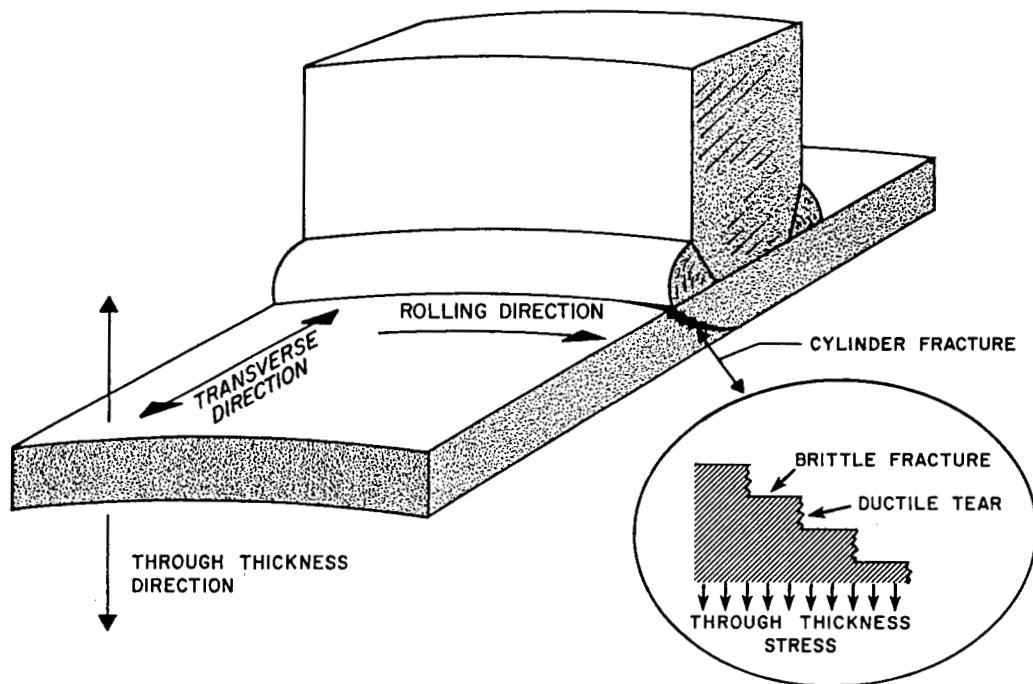


Fig. 3. Location in Cylinder Fracture and Rolling
Orientation of Shell Plate

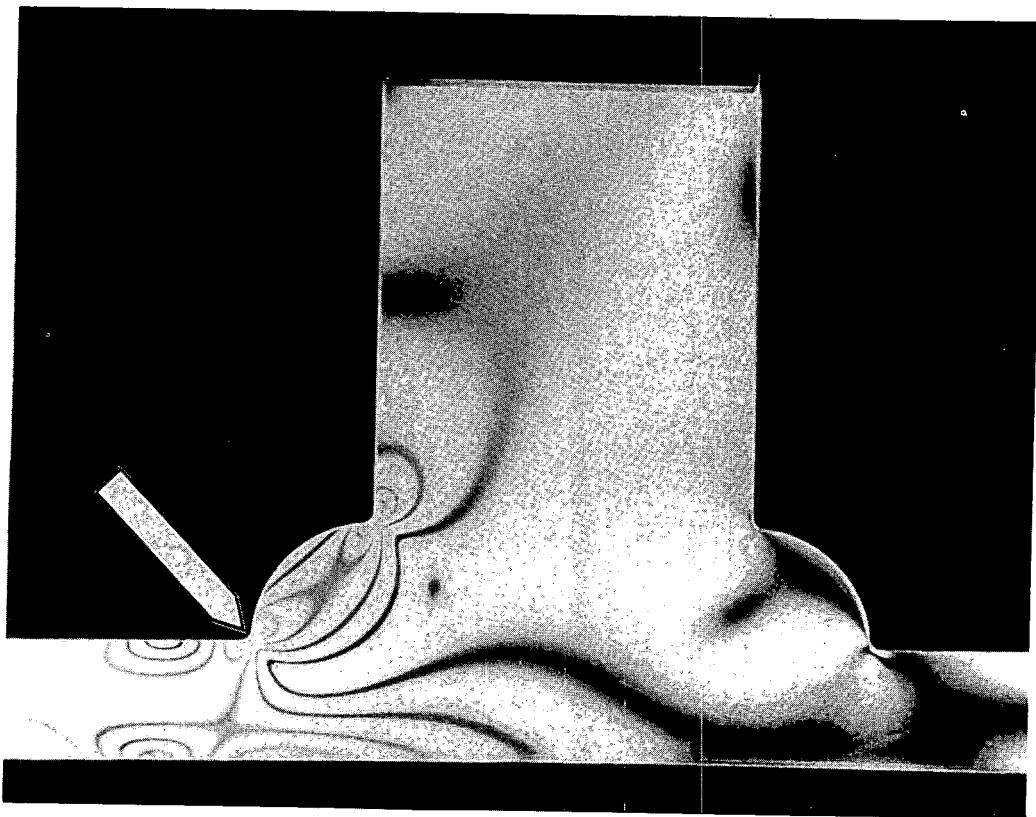


Fig. 4. Photograph of the Fringe Pattern of a Two-Dimensional Photoelastic Model of Stiffening Ring Weldment with a Convex Profile

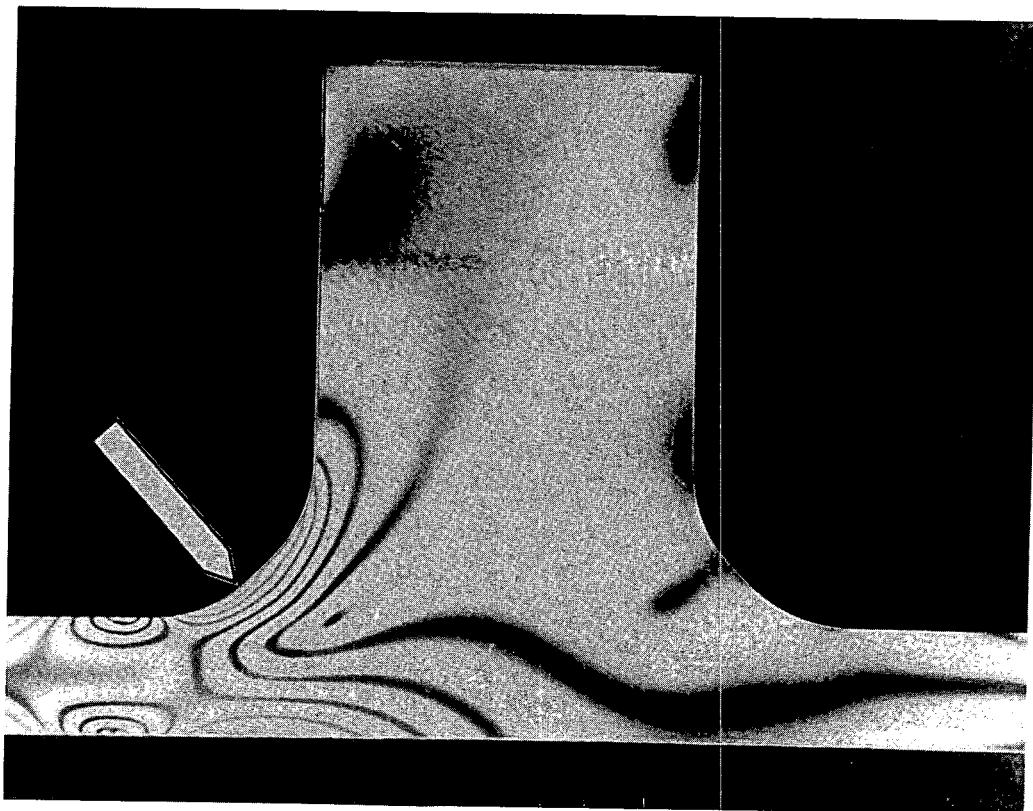


Fig. 5. Model of a Concave Fillet Profile

NOTE: TEST SPECIMENS FROM THIN WALL CYLINDERS, 5/16" TEST PLATES.
(IMPACT STRENGTHS BASED ON FULL SIZE 10 mm SPECIMENS)

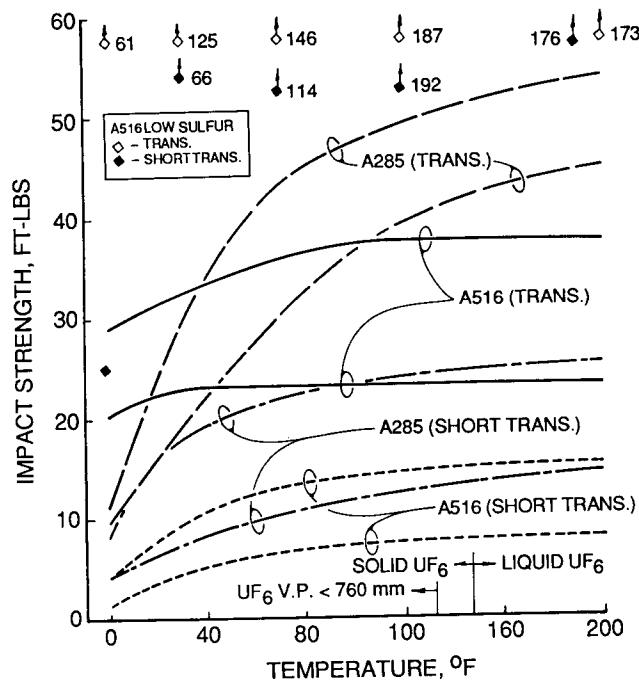


Fig. 6. Temperature Dependence of Impact Strength of Cylinder Steels

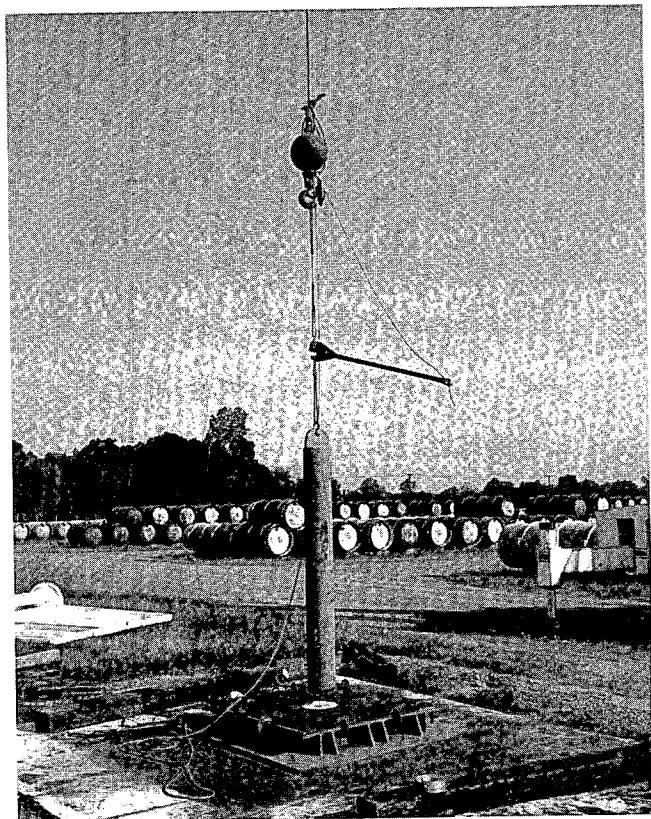


Fig. 8. Drop Test System to Measure Fracture Resistance of Stiffened Steel Plates

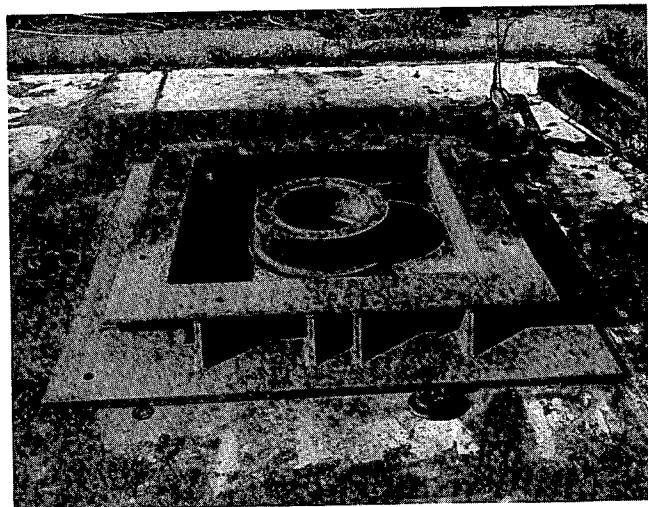


Fig. 7. Frame for Holding Stiffened Test Plates

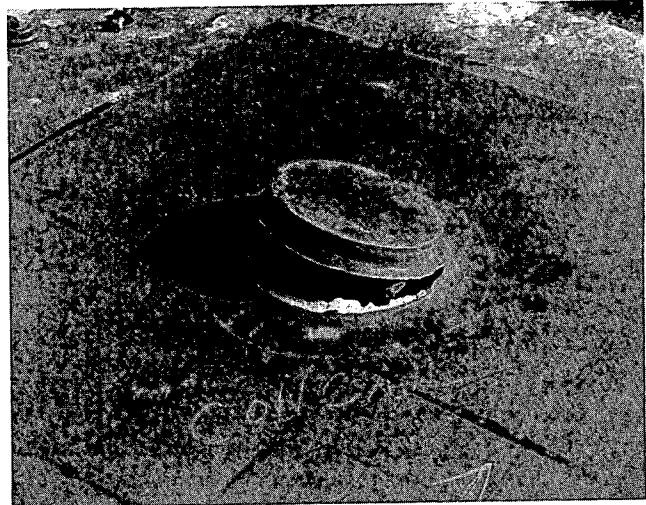


Fig. 9. Test Plate Fracture After Impact by Free Falling 1000-Pound Weight

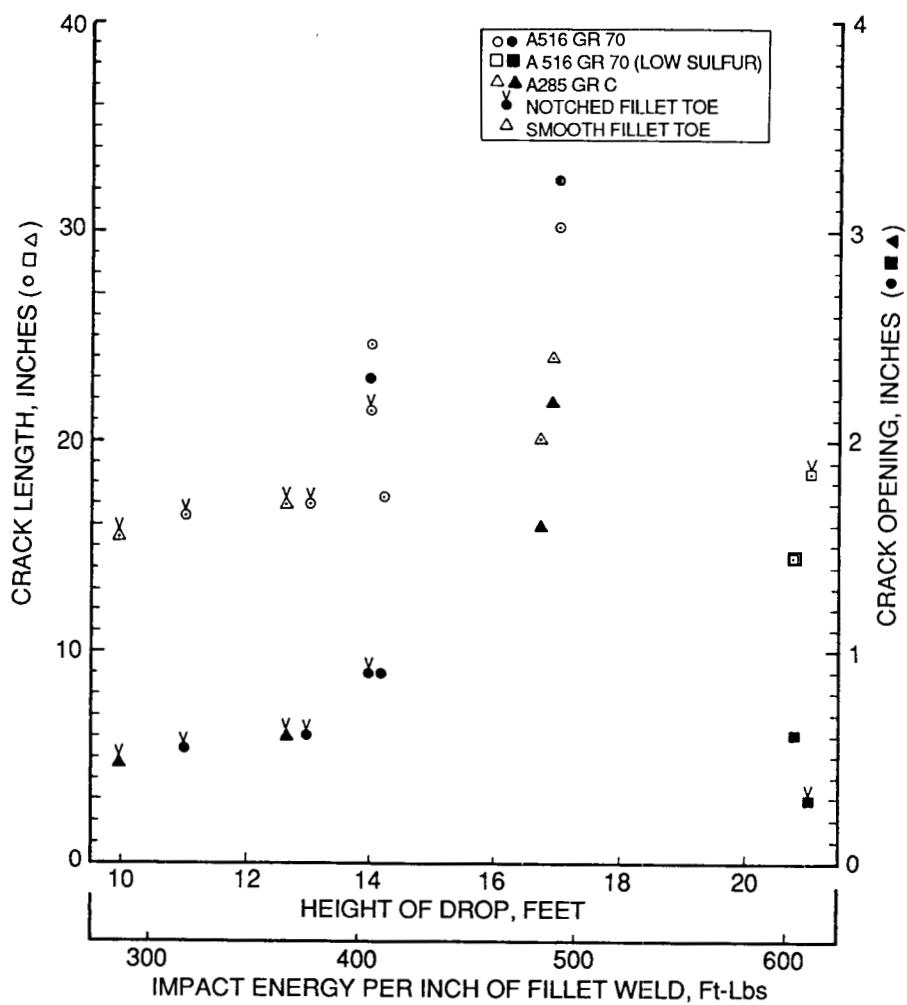


Fig. 10. The Effect of Steel Type and Weld on Propagation of Cracks in Stiffened Steel Plates

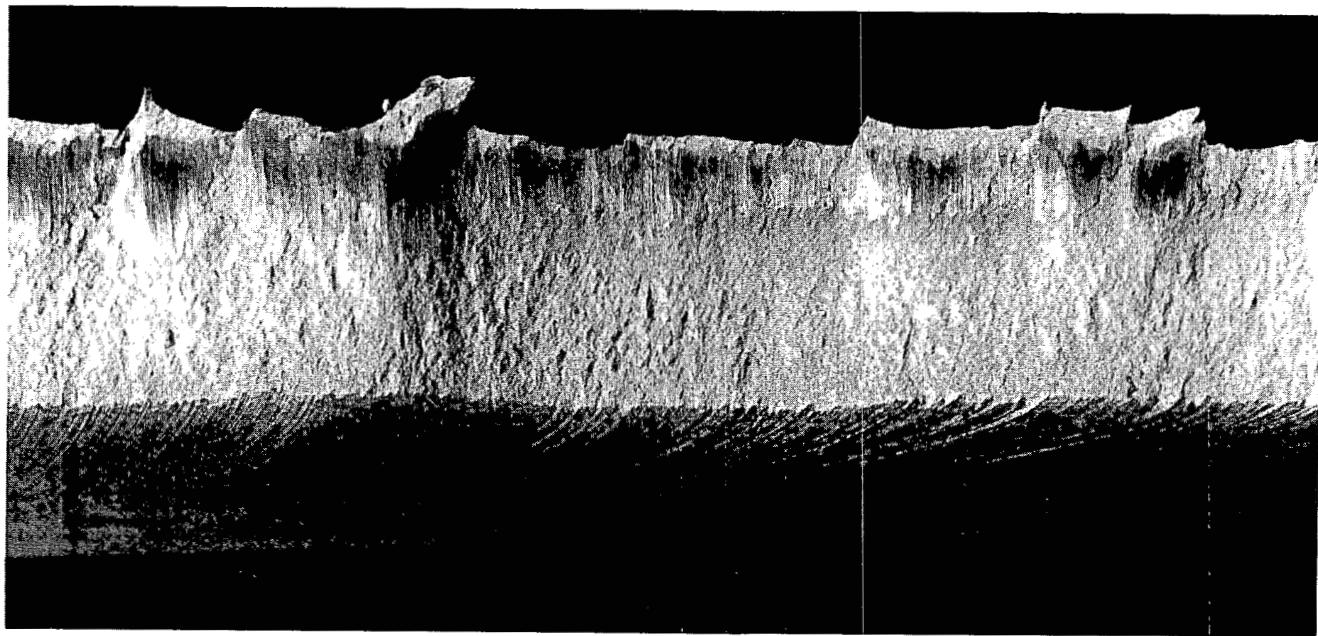


Fig. 11. Fracture Face of Ductilely Torn A516 Plate Made by Low Sulfur Practice with Inclusion Shape Control

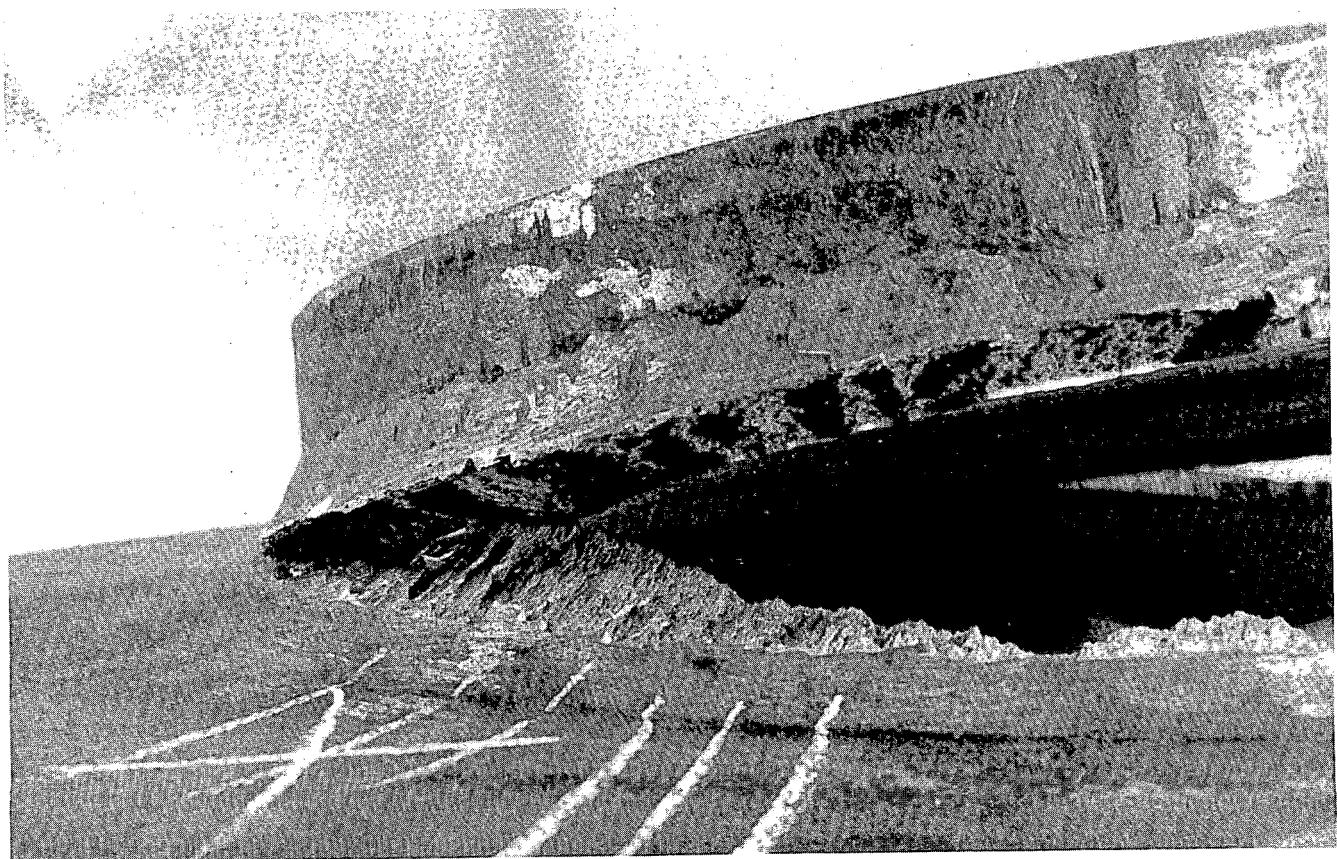


Fig. 12. Lamellar Tearing of A516 Steel Plate

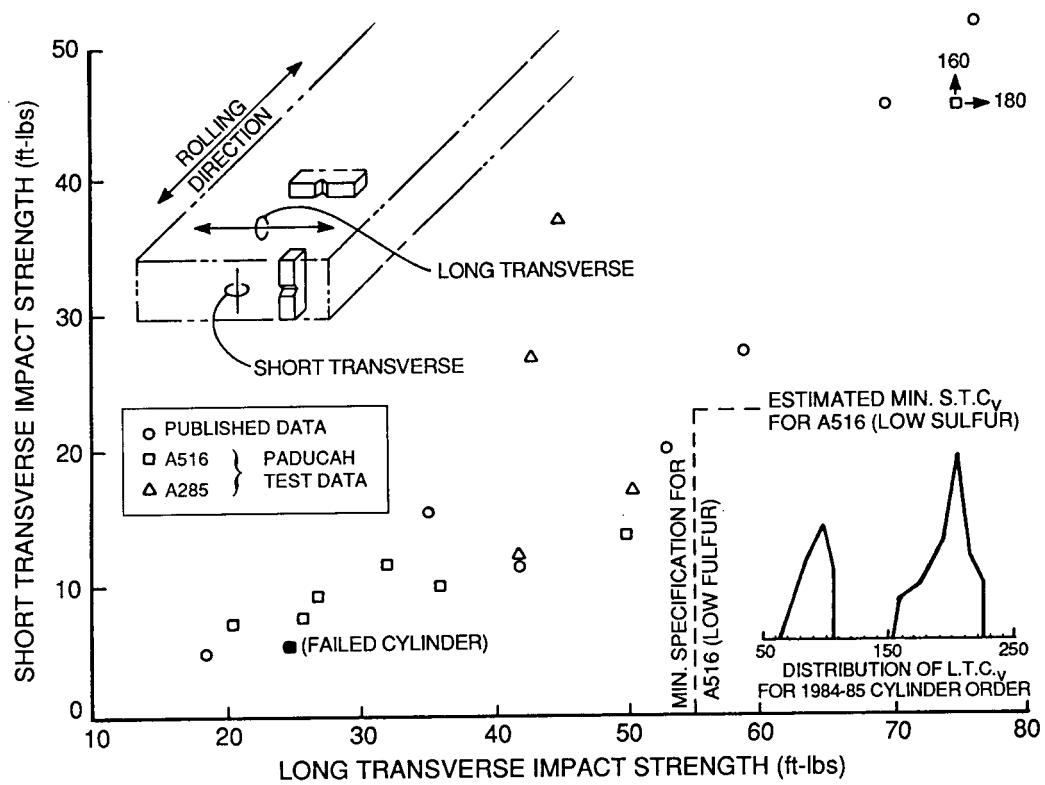


Fig. 13. Orientation Effect on Charpy V-Notch Impact Strength

THERMAL TESTS ON UF 6 CONTAINERS AND VALVES
MODELISATION AND EXTRAPOLATION ON REAL FIRE SITUATIONS.

B. DURET, P. WARNIEZ

ABSTRACT

From realistic tests on containers or on valves, we propose a modelisation which we apply to 3 particular problems :

- Resistance of a 48 Y containers, during a fire situation.
- Influence of the presence of a valve.
- Evaluation of a leakage through a breach, mechanically created before a fire.

I. INTRODUCTION

The resistance evaluation to fire of the UF 6 container, type 30 B or 48 Y, without a protection shell, is the principal matter of our study. A préliminary work had been the purpose of a communication to the PATRAM 83 [1]. We are continuing with this work in the present note, by improving the anticipation of the behaviour of a package supposed to be impervious, but also, by supposing a leakage at the valve and in the case of a breach mechanically created before a fire.

II. INTACT CONTAINER AND TIGHT VALVE

Because the physical characteristics of the UF 6 cannot be simulated in a satisfying way, we have to base ourselves on tests done with UF 6.

In the case of a generalized fire applied to a container without a shell, we only have tests conducted in 1965 [2]. The experiment consisted in measuring the exposure time to a hydrocarbide fire, until rupture of the small containers (maximal mass 113 kg for UF 6 against 2.3 tons for 30 B and 12.6 tons for 48 Y).

For two reasons, the interpretation of these tests can be considered as partial :

- Unknowledgement of the type, of the thermal level and of spreading in space of the external heat flow.
- Lack of instrumentation in the container.

Other heating experiences exist, for example, a 10 tons instrumented container [3] or the experience conducted by the PRNFDC [4], but in both cases, the heating temperature, never exceeds, respectively 93°C and 400°C.

II.1. Modelisation of physical phenomena

- a) External received heat flow in a container during a fire :

The AIEA reglementations, for type A packages (which

does not at all correspond to the classification of a 48 Y or 30 B package) advises to suppose that the temperature is equal to 800°C and by choosing emissivities of .9 for the flames and .8 for the envelope of the container. This means that the received heat flow depends on the steel temperature, therefore, on the internal thermal flow between the envelope and the UF 6.

Therefore, at a 20°C temperature, the heat flux will be 54 000 W/m² and, for example, if the steel temperature is 500°C the heat flux will be 40 000 W/m². In addition to the radiative heat flow, must be added a part of natural convection (10 % of the radiant flow) and of forced convection heat transport.

A recente note by F. NITSHE [5] indicates that, during a hydrocarbide fire around some small containers, the heat flow value is of 25 W/m²°C. The real heat flow, depends on the container's size compared with the size of the fire ; we can base on tests carried out in France, with a crude oil fire, covering a surface of 2000 m² [6] : it appeared, that the flame temperature decreases fast with altitude (which is maximum of 1 meter) and may reach 1250°C, an average radiance of the flames has been estimated at 30 000 W/m².

Depending on the case studied, (realistic calculation or pessimiste risk estimation) we can choose one of the previously described cases, as to introduce it as an incoming parameter for the calculations.

b) Internal transfers in the container

Principal physical phenomena which may have an influence :

- Conduction inside the steel lining
 - . well mastered (in spite of the lack of precise data on λ, ρ, C_p of the ordinary steel, as a function of the temperature).
- Thermal resistance between steel and UF 6
 - . depends on the type of filling (liquid ou gaseous)
- Thermal transfer internal to the UF 6
 - . Radiation : knowledge of an emissivity of the UF6 in a solid state
 - . conduction : thermal problems with the circulation of UF 6 (solid, liquid, gaseous)
 - . Effect of a triple point at a temperature of 64°C
 - . Influence of UF 6 gas (at 54°C the vapour pressure above solid UF 6, equals 1 Bar) on the transfer of the mass inside the cracks (heat pipe effect).

- Unknown convection
- Unknown boiling
- Critical point at 230°C. State equation.

In addition to these phenomena, we must add some internal mass transfers, due to the important vapour pressures (70°C 1.8 Bar, 100°C 4.1 Bars and 120°C 6.5 Bars) ; such transfers may considerably increase the apparent thermal conductivity ; it may then, be multiplied by a factor 10, between ambient temperatures and 65°C [1].

Our work is actually leading us to propose an internal heat transfer model), which has the following principal characteristics :

- Transient conduction inside the steel envelope
- Radiation heat transfer and conduction to the interface steel - Solid UF 6
- Apparent good conductibility in the UF 6, allowing for homogeneous temperature
- Increase of heat transfers while changing phases, from solid to liquid : transfer by boiling after phase changing at 64°C.

c) Rupture

Rupture happens if the internal pressure involves an over-strain of the rupture limit of the steel or if the liquid UF 6 fills all the available volume. This is a hydraulic rupture.

It appears that, in the case of the UF 6 thermal model that we have chosen, the hydraulic rupture appears before the mechanical rupture.

II.2. Application to real tests

The interpretation of the Japanese experiments [4] on 110 Kg of UF 6 containers electrically heated permits to verify our model of heat transfer in the UF 6.

Main characteristics of a test at 400°C :

- Heating by radiation of electrical resistors
- Good knowledge of the STPT 38 steel temperatures
- Temperature measurements in the UF 6.

The application of our model has been accomplished by using a one dimensional transient approach.

The table 1 resumes the principal date used for the calculation.

	<u>Table 1</u>	SI Value
Geometry :	External diameter	.267
	Steel thickness	.0286
	External surface	1.286
	Internal volume	
Steel :	Thermal conductivity	55
	Density	7850
	Heating capacity	490
	Coefficient of thermal expansion	$1.2 \cdot 10^{-5}$
	Density at 20°C	5090
	Density at 64°C (solid)	4920
UF 6	Density (liquid) $4130-7.13 t$ (t in °C)	
	Heat capacity (solid)	487
	Heating capacity (liquid)	558
	Gas thermal conductivity	.007
	Fusion heat at 64°C	54480

The heat exchange between steel and UF 6 is controlled through a gas film (the initial thickness is 0.9 mm); choosing an external emissivity of 0.6, which includes the radiation effect and a part of the convection linked to surrounding air, we then obtain, a steel temperature equal to 96°C after 20 minutes and 111°C at the 30th minute, the inter-

nal exchange by radiation, with an emissivity of 0.3 is equivalent to the exchange by conduction through the film ; about 700 W/m².

A calculation compared to a test is shown in fig.1. Steel temperature falls, when the UF 6 joins the melting point which is obtained at the 33 rd minute, then appears an homogeneisation of the temperatures between steel and UF 6, the temperature is under estimated at 72°C, which means that the external heat flow, during the heating period, may be more important.

When is noticed, on the one hand, that the steel temperature does not exceed 120°C, and on the other hand, that the diameter of the container is only 1/6 th of a 48 Y. Then we will come to the conclusion that extrapolation of this model, on a big UF 6 container during a fire at a temperature of 800°C or 1000°C, is hazardous.

Nevertheless, by calculating the tests conducted by MALETT [2] (the steel has arrived at a temperature of 540°C during the test) we obtain a rupture after around 8 minutes if we suppose an external average emissivity of .7. This time is experimentally verified.

II.3. EXTRAPOLATION ON A FIRE AROUND A 48 Y

Supposing an internal heat transfer model, identical to the previous case, we study the time until rupture, with two assumptions :

- First case : maximum external flow with a temperature of 800°C and an emissivity Fire-Steel of .9 and .8, respectively adding a natural convection flow around the cylinder.
- Second case : more realistic external flow, with a value of 30 kW/m².

On the other hand we suppose that all the external surface receives the heat flow.

In fig. 2 we give the transient temperature histories between steel and UF 6, in both cas. By calculating the steel temperatures, which are important (above 500°C), we obtain internal radiative heat flows, which are 10 to 30 times above the heat flow due to conduction through the gas film.

In the first case, the hydraulic rupture (T of UF6 148°C) happens after 41 minutes and 57 minutes in the second case.

III. LOSS OF TIGHTNESS AT A VALVE

UF 6 industrial containers (30 B or 48 Y) are equipped with a valve and a plug. For the following reasons, we think that such accessories seem to be specially sensible, in the case of a fire :

- 1) The tightness of the valves and of the plug is obtained by using a layer of alloy which is applied on their screwing points. Lead-tin alloy melts at 200°C.
- 2) The valves are made with materials having different thermal dilatation coefficients. For example the body is made of aluminium and the rod is made of monel (See fig. n° 3).

III.1. Experiment

The experimental system, sketched on fig. 4, allows a progressive or a sudden heating of a complete lot of valves.

The electrical oven, equipped with a steel tube used as a protection in case of a rupture of the valve, creates a hot surrounding (800°C).

In order to avoid an eventual oxidization, we have chosen nitrogen for the tests. A metallic container

fixed on the support, with an interposed gasket is used to receive the valve leakage flow. The gas is then conducted to nitrogen-water heat exchanges permitting to lower the nitrogen temperature to its normal temperature before passing in a flow meter

a) Progressive heating :

The temperature measurements and the leakage measurements are conducted by imposing the oven temperatures by steps (100, 200, 300, 400°C) and then by varying the pressure (5, 10, 20 and 30 bars). Five valves have been tested, (see recording in fig. 5). The melting of the alloy is evident, nevertheless, we have noticed the beginning of a leakage at 100-150°C, which seems to indicate that a leakage, can be initiated by a differential dilatation of valve elements, even more important leakages that the thermal gradient, internal to the valve, is important.

b) Sudden heating :

All the devices are the same, but the valve and the plug are screwed on a sleeve which is welded same measurement thermocouples are in place.

The experiment consists to heat the oven, which is suspended above the valve, until obtention of a steady temperature (800°C) inside the steel tube and bring the oven over the valve, which is maintained at a pressure of 5 bars. The valve temperature increases, then we can note the time it takes and the temperature level of the surrounding space at the beginning of the leakage.

If the gasket, made of PTFE (see fig.3) does not exist, a leakage appears after 1 minute and 25 seconds, when the scriwing is at a temperature of 78°C.

For a completely equipped valve, the leakage starts when the temperature is of 200°C (melting temperature of the lead-tin alloy) after 4 minutes 50 seconds.

III.2. Extrapolation on gaseous UF 6

If the valve has lost its tightness, the leakage depends, of the motor pressure, or during a fire, the leakage depends on the obtained temperature, which depends on internal thermal flow to the UF 6. Supposing that the UF 6 temperature is of 140°C, before a hydraulic rupture, the pressure would cause a leakage of about 2 N.m³/h of nitrogen in the experiment .

Supposing adiabatic behaviour of the UF 6, then :

$$Q_{UF6} = Q_{Nitrogen} \frac{MN_2}{MUF6} \frac{2 \gamma UF6}{\gamma UF6 + 1} \frac{2 \gamma N_2}{\gamma N_2 + 1}$$

Based on the previous choices $Q_{UF6} = 0.55 \text{ m}^3/\text{h}$ which means that after hydrolysing we have an emanation of 0.5 g/s in HF.

container emissivity of .8 we considered also a contribution of natural convection.

- Second case : the heat flow is 30 000 W/m² and the fire lasts 15 minutes (see results in fig. 6 and 7) In any case, the steel envelope exits and has an influence by its thermal inertia ; the heating continues up to the time at which, an external intervention permits a fast cooling of the steel.

In the following table we present the main results:

Table 2

	UF6 Density evacuated (Tons)	
	if intervention	without intervention
First case 800°C duration : 30 minutes	3.44	5.9
Second case 30 kW/m ² duration : 15 minutes	1.1	2.7

If the breach is small enough, the pressure drop, and a shrinking related to a UF 6 deposition, can decrease considerably the leakage flow.

V. CONCLUSION

The uncertainty of our evaluations resides on two important points.

1. External heat exchange : it is variable because of the homogeneity of the flames and the smoke influence, and the modification of the container's emissivity. A modelisation is utopic because each fire is a different case, which depends on :

- the type of hydrocarbide
- the exact position of the container

We think that this exchange must be an incoming parameter of the calculation program, knowing that it can involve variations on the rupture time, for example around 50 %.

2. Steel-UF 6 heat transfers

Because we actually, don't have high temperature heating experiments with big diameter UF 6 containers, we cannot give a definition of the moments when ruptures will happen on industrial containers, as the estimations can vary between 15 minutes and 1 hour on different models, but joining the rupture time, in the MALLET tests [2].

Different assumptions are possible :

- a) Considering the important UF 6 density, the heating can stay limited to the peripheral zones generating some growing pressures which are localised at the periphery.
- b) A part of the UF 6 can be liquefied and would be accumulated at the bottom of the container, then two possibilities appears :
 - The container is involved intirely by fire, including the bottom and involves an important spraying of UF 6, leading to a global heating with an excellent heat transfer and an early rupture of the envelope.
 - The fire does not reach the liquid zone, and the existence of a bad coefficient of the heat transfer between the lining and the solid UF 6 delays the UF 6 heating and the rupture of the container.

The remaining problem is to determine the right manner, the conditions of phase changements, the mass transfers inside the container which would determine the conditions of the rupture time, as well as the quantities of UF₆ which are susceptible to be released if there is a leakage at the valve.

Actually a test is envisaged. It's principle is the following :

- Transient heating of a 1.3 m portion of a container 48 Y correctly instrumented, internally and on the shell surface.
- Good knowledge of the external transient heat flow at a thermal fire level of 800 to 900°C.
- Stopping the test before rupture, but with an oven situated in a spacious surrounding, allowing a release of the UF₆.

The main parameters must be :

- The rate and the type of filling (gaseous or liquid),
- protection or not of the valve (leakage measurements),
- Oven temperature,
- Heating duration.

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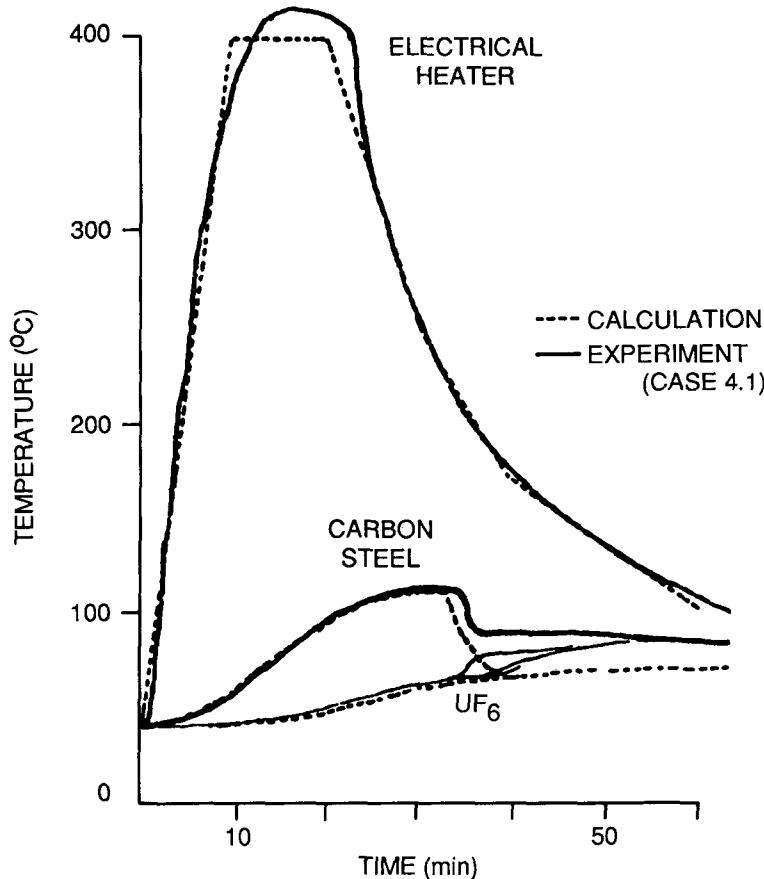


Fig. 1. Model comparison with Japanese experiment (4)

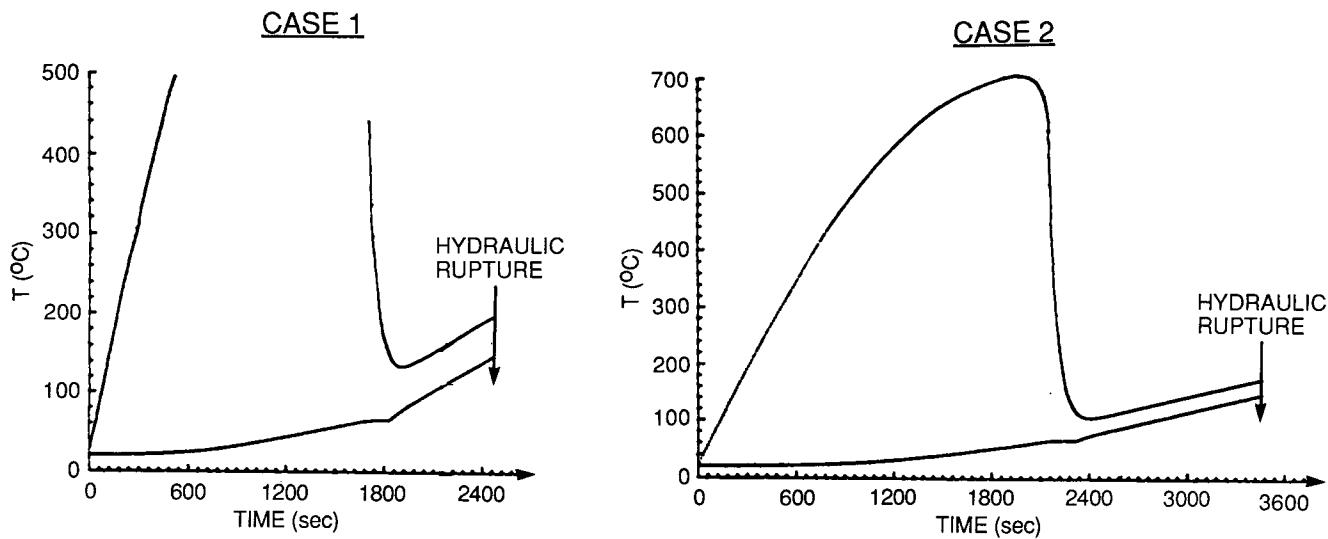


Fig. 2. Extrapolation to real fire on 48Y container

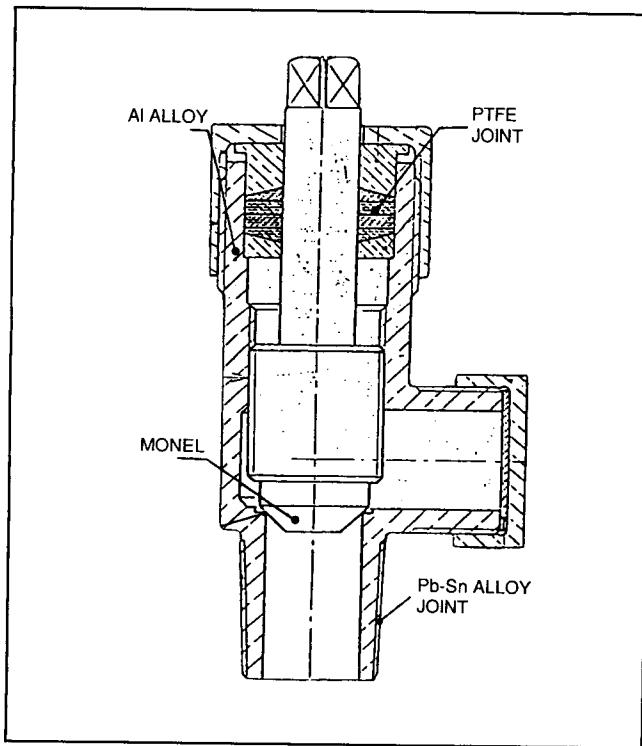


Fig. 3. 48Y valve schematic

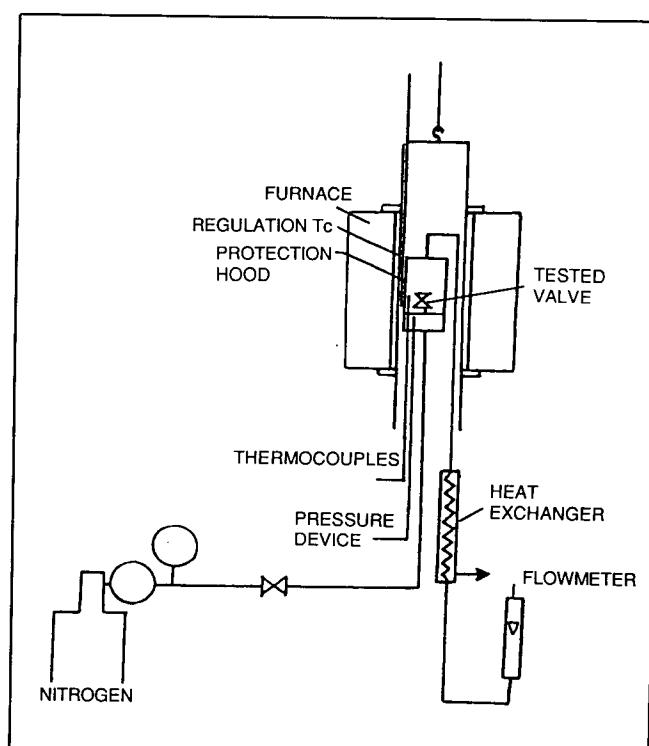


Fig. 4. Valve test loop

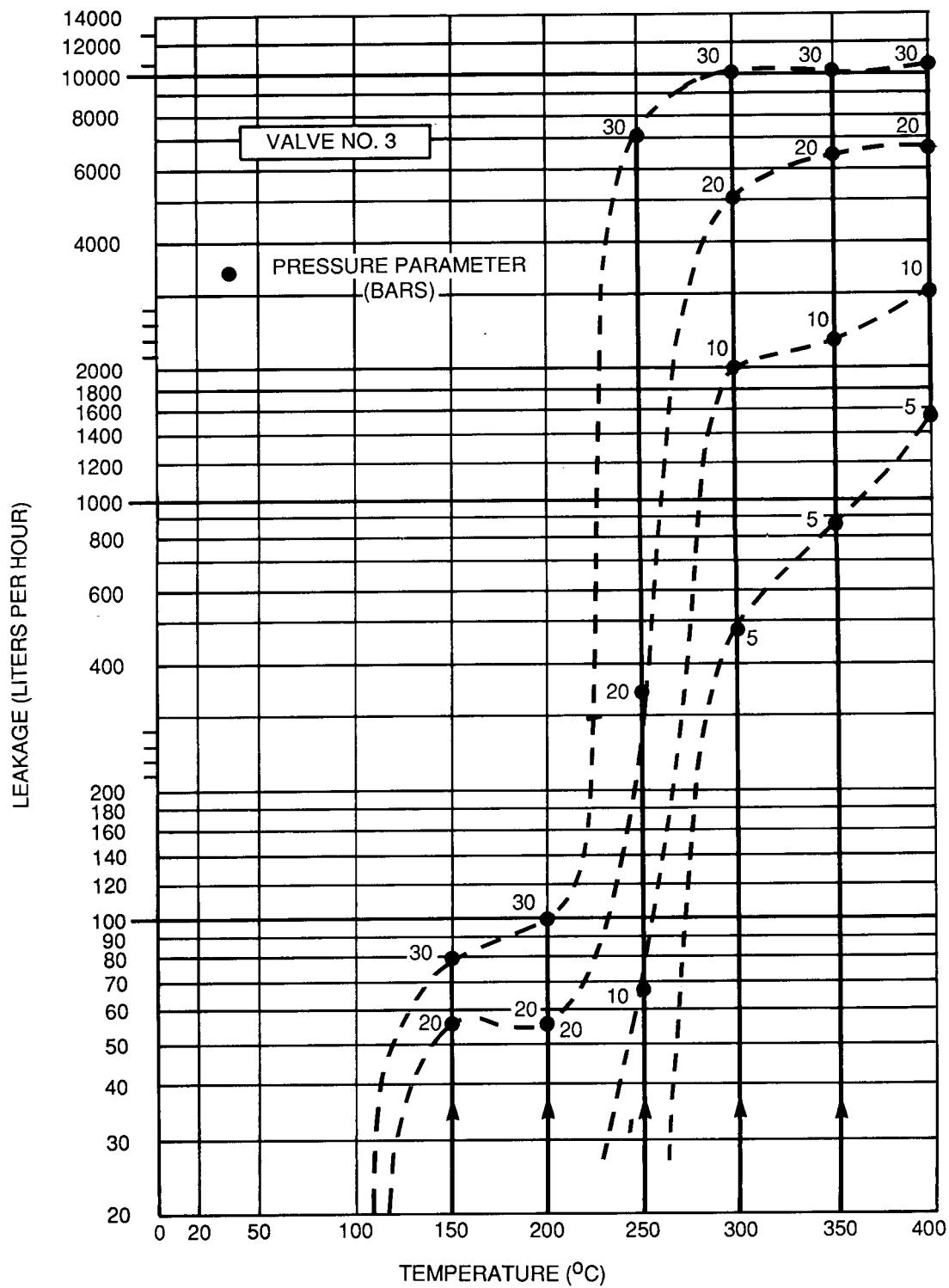


Fig. 5. Experiment measurements

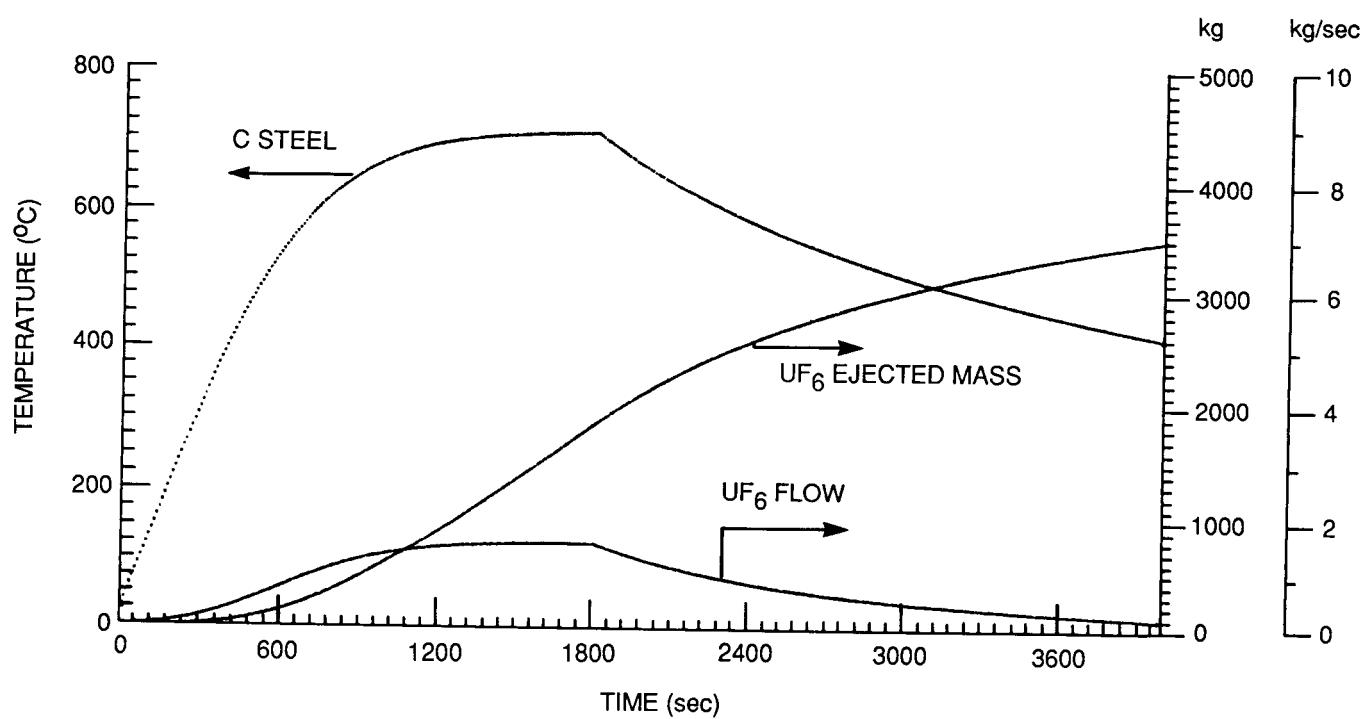


Fig. 6. Case 1 (fire duration = 30 min)

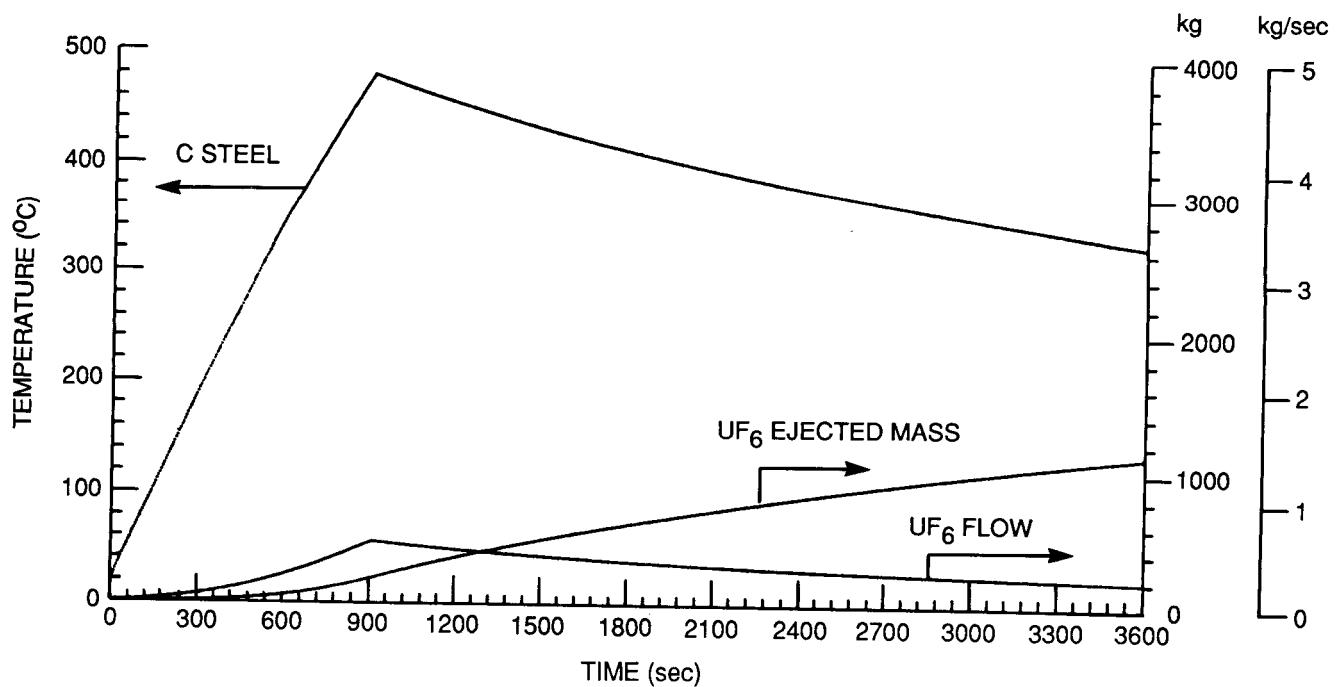


Fig. 7. Case 2 (fire duration = 15 min)

*RUPTURE TESTING OF UF₆ TRANSPORT AND STORAGE CYLINDERS

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ABSTRACT

Rupture tests have been conducted on pressure vessels of all the sizes and designs in current commercial use in the transport and storage of uranium hexafluoride. The test results have demonstrated the general conformance of the cylinders to their design criteria. Several of the test cylinders were taken from active service. A few were made available as the result of handling mishaps: These were a dented 48-inch cylinder and two type 30B cylinders damaged in a warehouse fire. Some cylinders were tested to evaluate the effects of modifications or repairs, including 5-inch cylinders with new valve couplings and 12-inch cylinders modified for use as cold traps. The repair and modification welds had no effect on rupture strength or failure location; therefore, all the test results are viewed as representative of the cylinders currently in use for shipping and storage.

The tests have shown that the stiffening ring butt weld serves to localize the failure of the 48-inch cylinders, with an associated penalty in ultimate pressure. They have also shown the extreme conservatism of design in the case of the 5-inch cylinder, the only product cylinder qualified for 100% enriched material. Finally, they have shown a basic tendency toward brittle fracture in over-pressuring of the DOT-qualified type 30A cylinder, resulting in a recommendation for discontinuation of its use as a transport cylinder.

*Based on work performed at the Oak Ridge Gaseous Diffusion Plant operated by Martin Marietta Energy Systems, Inc. for the U. S. Department of Energy under contract DE-AC05-84OR21400.

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INTRODUCTION

Over 50,000 cylinders for transport or storage of uranium hexafluoride have been purchased by the gaseous diffusion plants since 1950 and most are still within the DOE-ORO complex, Table I. Most were built to criteria established by the ASME Boiler and Pressure Vessel Code. Aside from small sample cylinders, these cylinders range from 5 to 48 inches in diameter and are sized to contain from 50 pounds to 14 tons of uranium hexafluoride at assay levels ranging from depleted tails to 100% enrichment.

Table I
DOE UF₆ CYLINDER PROCUREMENT HISTORY

Type of Cylinder	Description	Number Purchased	Procurement Dates
1S	1 inch	217	57-76
2S	3 inch	1,841	53-81
5A	5 inch	1,009	53-64
5B	5 inch	100	1984
8A	8 inch	236	55-65
12	12 inch	318	55-65
30A	2.5 ton	450	50-60
30B	2.5 ton	122	1986
48A	10 ton	1,365	51-54
48T	*10 ton	4,230	56-58
48X	10 ton	1,500	53-54
480	*14 ton	6,602	58-61
480M	*14 ton	16,370	62-78
48G	*14 ton	11,900	73-86
48F	14 ton	90	61-62
48Y	14 ton	260	79-80
48HX	*14 ton	1,000	1979
48H	*14 ton	3,140	79-86
*Thin Wall		TOTAL-50,750	

The cylinders intended for long-term storage of depleted process tails are primarily 48-inch diameter cylinders designed for working pressures of 100 psig. These have capacities of 10 and 14 tons and have a nominal wall thickness of 5/16 inch. Cylinders of similar UF₆ capacity, which were designed for transport of UF₆ as feed material and as low-enrichment product, have a wall thickness of 5/8 inch and are nameplate-rated for 200 psig working pressure. The 48-inch cylinders have been procured in a number of design modifications involving primarily external fittings and hardware. The pressure envelope, however, is similar for all these designs, with the two wall thicknesses distinguishing tails storage cylinders from those intended for shipment or transfer of feed or product materials. These cylinders are made from mild steels conforming to ASTM plate specifications A516 or A285, depending on procurement date. Strength levels for these steels are in the 55,000 to 70,000 psi range. All were manufactured as Code vessels.

Steel product cylinders of 2-1/2 ton capacity have been manufactured in two configurations, designated 30A and 30B. The 30B cylinder, with a 200 psig working pressure designation, is of conventional pressure vessel configuration and was manufactured as a Code vessel. The 30A cylinder, similar to the standard 1-ton chlorine cylinder, was manufactured to DOT specifications and consists of a seam-welded right circular cylinder with concave heads attached by forge welding. Its working pressure is 250 psig. The design has been declared obsolete for UF₆ service in favor of the 30B design, but there are over 3,000 such cylinders on hand, most manufactured prior to 1945 as chlorine cylinders and subsequently modified for UF₆ use. Some were manufactured in the 1950 to 1960 period, and an additional small lot was manufactured in 1984. About 1,000 are in use in storage of UF₆ material, and some still serve in transport of enriched product material.

The smaller product cylinders, all rated for working pressures of 200 psig, are made from nickel or Monel. Two 12-inch, 460-pound capacity cylinders are in use - a spun, seamless nickel cylinder, and a seam-welded Monel cylinder. The 8-inch, 265-pound cylinder, made from seam-welded sheet and formed ellipsoidal heads, is made from either nickel or Monel, as is the 5-inch model of 55-pound capacity which is fabricated from standard pipe and weld caps. All of the nickel and Monel cylinders were fabricated as Code-compliant vessels, although weld modifications made on many of the 12-inch nickel cylinders and most of the 5-inch Monel cylinders have invalidated the code classifications which initially applied.

The design considerations in the ASME Code specify a maximum stress in the cylinder wall which generally identifies the maximum hydrostatic test pressure and, hence, the working pressure (half of the hydrostatic test pressure for this group of UF₆ cylinders). Where the wall thickness is selected based on these considerations, the burst pressure will

generally be about eight to ten times the working pressure. Notable exceptions in the present group of cylinders are the 30A cylinder which, as earlier noted, is not an ASME design, and the 5-inch cylinders which, although built as Code vessels, develop only a small fraction of the allowable stress at the specified test pressure.

At various times over the past 25-30 years, design and safety studies related to UF₆ packaging for storage and transport have been supported in part by burst testing of UF₆ cylinders. These tests have covered most of the cylinder varieties in present use in the uranium fuel cycle, including the two exceptions to the standard designs noted above. This report presents the test data accumulated in recent tests of large product and storage cylinders, tests of weld-modified 5-inch and 12-inch cylinders, tests on a pair of fire-exposed Model 30B cylinders, tests on two Model 30A units, and tests on 5-, 8-, and 12-inch cylinders in support of a packaging development program. Some of the earlier tests were covered in individual reports or internal memoranda; tests on the large cylinders have not been previously reported.

SUMMARY

Nickel and Monel product cylinders in 8-inch and 12-inch sizes gave burst pressures of 12 to 15 times the nameplate working pressure. The 5-inch cylinder, manufactured from schedule 40 pipe and developing only a small fraction of the allowable stress at the specified hydrostatic test pressure, proved to be an extremely conservative design and failed at 40 times the rated working pressure.

The Model 30B steel cylinder, of similar design and configuration, gave burst pressures 11.3 and 11.8 times the 200 psig working pressure. The design - obsolete 30A cylinders, with a working pressure of 250 psig, showed sudden head reversal at 900-1,000 psig and (brittle) failure by head separation at 1,250 psig for a safety factor of only 5.0.

The large (48-inch) shipping and storage cylinders, of conventional design but with added external stiffening rings, suffered some strength reductions and lowered safety factors because of end-joint failures in the stiffening rings during the burst tests. Safety factors of 8.8, 8.7, and 6.4 were observed in tests on one thin-wall storage cylinder and two heavy-wall feed cylinders.

PROCEDURE

In general, the test cylinders were obtained from active service for burst testing. In some cases, they had been damaged in handling accidents and did not qualify for further service without detailed examination, evaluation, and repair of the damage. Two 30B cylinders were obtained from a transport services firm in the feed enrichment cycle following their exposure to a warehouse fire which subjected them to temperatures as high as

Table II.
RUPTURE TEST DATA FOR UF₆ CYLINDERS

Cylinder Type	Material	Wall Thickness Inch	Working Pressure PSI	Rupture Pressure PSI	Percent Volume Increase
5A	Monel	1/4	200	8250	
5A	Monel, Nickel Coupling	1/4	200	7950	21
8A	Monel	3/16	200	2950	
8B	Nickel	3/16	200	2450	
12A	Nickel	0.200	200	2400	30
12B	Monel	0.250	200	2260	53
30A	Steel (A285)	13/32	250	1250	20*
30B	Steel (A516)	1/2	200	2270,2360	34
480M	Steel (A285)	5/16	100	870	9
48Y	Steel (A516)	5/8	200	1770	6
48A	Steel (A285)	5/8	200	1285	6

* The volume increase of the 30A cylinder is due to inversion of the concave heads at an internal pressure of 900 - 1,000 psi.

1600°F. One 48-inch cylinder, never in service, was dented and thus not suitable for filling. Three 12-inch cylinders were tested from a group of cylinders weld-modified for use as cold traps in UF₆ collection. A weld-modified 5-inch cylinder was tested to evaluate the effects of a material substitution in the cylinder head and valve coupling.

Where necessary, the cylinders were cleaned and decontaminated. In some cases, this included sandblasting the exterior surfaces so that strain gages could be attached to monitor the pressure tests. The cleaned, prepared cylinders were then fitted with new valves or special adapters, completely filled with water and connected to one of several high-pressure pumping systems. Pressure was monitored with a large, calibrated pressure gage; and, in addition, for the tests on the 48-inch and 5-inch cylinders by a transducer whose output was recorded periodically in a computerized data acquisition system. For the tests conducted prior to 1970, the cylinders were covered with heavy, woven rope explosion mats; later tests were conducted in a pit under a heavy steel cover. Failures in these latter tests were observed in a video monitor and some were videotaped to provide a permanent record.

The failures were photographed; and, in many cases, a volume was determined by measuring the cylinder distortion at several locations. Analysis of strain gage data gave information on cylinder response to pressure during the tests. Failure pressure data are presented in tabular

form, Table II. A photograph of a typical 48-inch cylinder failure (influenced by the premature failure of the end-weld joint in a stiffening ring) is also shown, Figure 1.

DISCUSSION

Most of the cylinder types in present use for transport of uranium hexafluoride are conservatively designed to criteria established by the ASME Boiler and Pressure Vessel Code, and burst tests conducted over a span of many years have demonstrated the safety against overpressuring inherent in these designs. Where wall thicknesses are selected on the basis of the Code maximum allowable stresses, the designs yield a minimum safety factor of eight times the specified working pressure, and conservatism in specification of minimum ultimate strength values for the selected materials gives, in practice, actual safety factors in the range of 10 to 15 times the working pressure. Examination of the test data shows this to be true of the 8- and 12-inch nickel and Monel product cylinders and of the 30B steel product cylinder. Design minimum safety factors of 8:1 are achieved in the 48-inch cylinders, but consistency in this respect is handicapped by the difficulty of assuring the specified full-penetration welds in the end joint in the external stiffening rings. In the three tests on 48-inch cylinders covered in this report, failure in all cases began in the stiffening ring joint and progressed through the attachment weld and into the pressure envelope at a net

stress well below the ultimate strength of the wall material. In the case of the 48A cylinder, the overall safety factor was less than seven times the working pressure; weld penetration at the stiffening ring weld was estimated at only 20%. Failure of the 48Y test cylinder was localized by the same mechanism; the net safety factor was 8.8:1.

The thin-wall 48.0M storage cylinder ruptured below a stiffening ring weld failure at a net safety factor of 8.7 times the (100 psig) working pressure. The flaw-dependent nature of the ruptures observed in 48-inch stiffening ring-reinforced cylinders indicates that the stiffening ring, including its end weld, can significantly alter the rupture characteristics of the cylinder. With full penetration end welds in the stiffening rings, the burst strength can be expected to rise substantially, and the failure location should move to an area between stiffening rings.

The 8-, 12-, and 48-inch cylinders, and the 30B cylinder, were all designed to ASME Code criteria. Selection of the required working pressure fixed the hydrostatic test pressure and, therefore, the maximum wall stress at that pressure. With adjustments for weld efficiency where required, the stress figure established the minimum wall thickness, and for seam-welded structures, the nearest higher standard commercial sheet or plate thickness was used for the cylinder wall. In the case of the 5-inch cylinders, however, it was considered more economical to use standard pipe and weld caps for ease in procurement and fabrication. Schedule 40 pipe and weld caps produced an orthodox cylinder configuration, and the material transfer process conditions were used to select the 200 psig working pressure. Stress calculations showed that the 2X hydrostatic test pressure would develop less than half of the Code maximum allowable stress in nickel and only one-third of the maximum allowable stress in Monel. This situation resulted in extremely high burst strengths in comparison to the larger cylinders, with safety factors in the range of 40 times the nameplate-specified working pressure. The fortuitously high safety factor is appropriate for this cylinder, however, considering the high intrinsic value of the contents in comparison to those in larger cylinders.

Over-pressure rupture of the DOT specification 30A cylinder is fundamentally different from that of the ASME code vessels. The concave heads invert suddenly at internal pressures which develop only about 60% of the ultimate strength in the walls of the cylinder. This inversion exerts a wedging action on the back side of the forge-welded closure, and subjects the adjacent wall section to plane strain conditions. The high loading rates associated with the head inversion initiate brittle cracks in the wall, and a moderate increase in pressure causes this cracking to propagate through the balance of the wall thickness. Hoop stress at this point is only 85% of the specified minimum strength of the steel used in the cylinder wall.

Where expansion of the cylinder contents is not limited (e.g., continued pumping or continued heating), the failure will involve complete separation of the end of the cylinder.

Because of the unconventional head attachment and the poor material utilization characteristics of the 30A design, it has been considered as obsolete since introduction of the 30B cylinder in 1970. The added drawback of a high risk of loss of the entire cylinder contents in the event of an over-pressuring accident is a cogent argument against continued procurement and continued utilization of the 30A cylinder design for transport of feed or product materials. Type 30A cylinders have been procured for UF₆ service as lately as 1984. The failure patterns shown in burst tests demonstrate an unacceptably high risk of release of cylinder contents in the event of accidental over-pressuring, and the ORO-651 recommendation in favor of the 30B cylinder should be followed in the interests of safety, economics, and environmental concern.

CONCLUSIONS

There are three principal observations to be made from the rupture testing of UF₆ cylinders conducted over the past 25 years:

1. The 30A cylinder (not designed to ASME Code criteria) is a basically unsafe design in the event of over-pressure, since the method of head attachment promotes brittle fracture which tends toward major, rather than minor, material releases.
2. Poor stiffening ring welds promote premature failure in burst testing of 48-inch cylinders (although most give the minimum Code safety factor of eight times the working pressure).
3. Cylinders for enriched product material which are designed to Code criteria give safety factors at rupture which are 11 to 15 times greater than the working pressure. The five-inch cylinder, for fully enriched material, had a rupture strength 40 times higher than the working pressure.

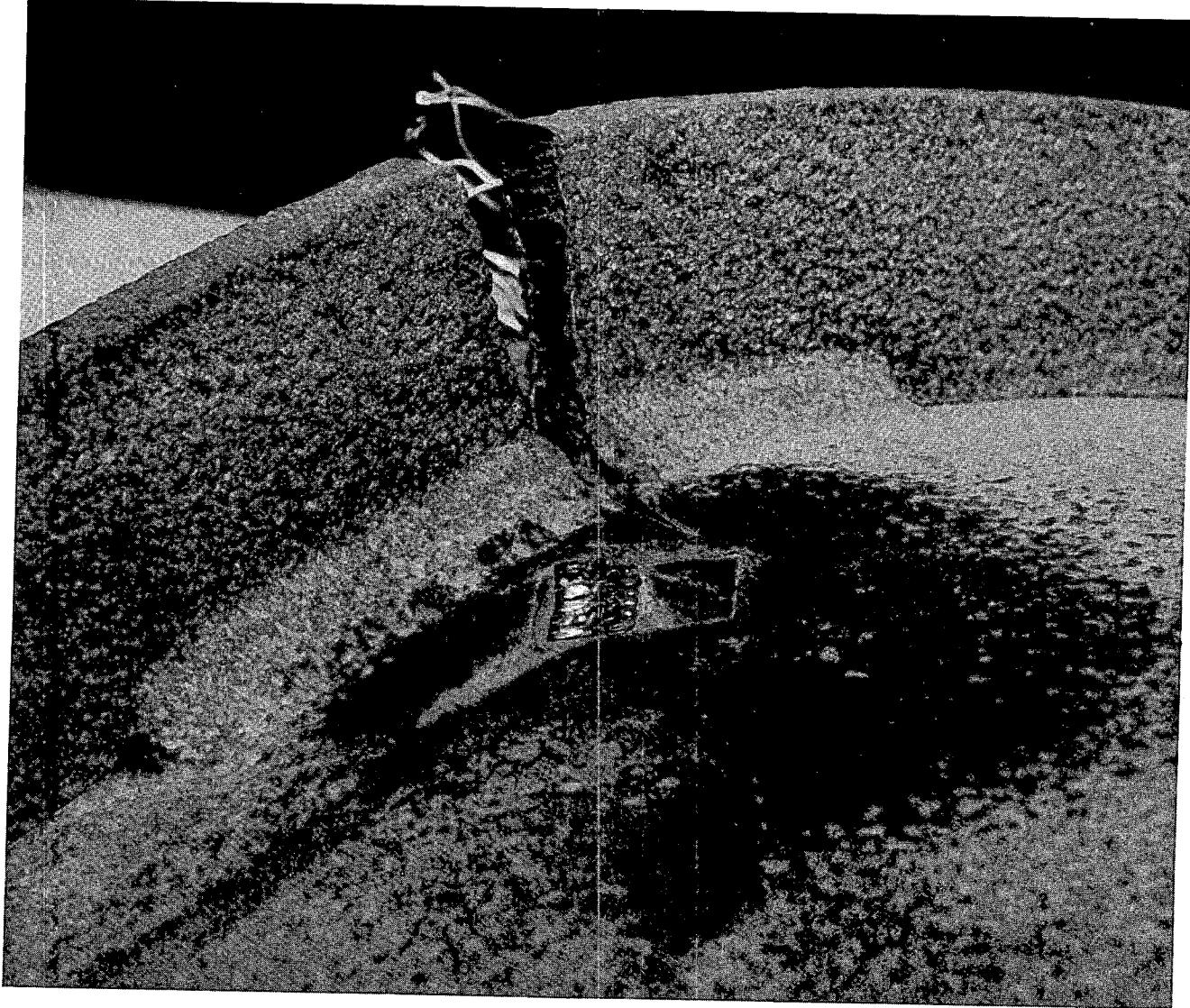


Figure 1.

RUPTURE AT STIFFENING RING JOINT IN 48Y CYLINDER

Fracture at the stiffening ring end closure joint localized the failure in the pressure envelope in all of the 48-inch cylinder tests.

101/02

MONITORING OF CORROSION IN ORGDP CYLINDER YARDS*

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ABSTRACT

Process tailings from U.S. uranium isotope enrichment activities are stored in mild steel cylinders designed and manufactured according to ASME Boiler and Pressure Vessel Code criteria. Most storage facilities are open areas adjacent to the enrichment plants where the cylinders are exposed to weather; approximately 5000 cylinders are in several cylinder yards at the Oak Ridge Gaseous Diffusion Plant (ORGDP). Since mild steel will corrode under these storage conditions, significant work is being done to determine general corrosion behavior of tails cylinders and to estimate anticipated lifetimes.

The program under way at the ORGDP is targeted at conditions specific to the Oak Ridge cylinder yards. The work includes (a) determination of the current conditions of cylinders stored in these yards, (b) description of rusting behavior in regions of the cylinders showing accelerated attack, (c) the monitoring of corrosion rates through periodic measurement of test coupons placed within the cylinder yards, and (d) establishment of a computer base to incorporate and retain these data.

The information obtained will enhance planning for continuing safe storage of the tails material.

INTRODUCTION

Uranium hexafluoride (UF_6) throughout the nuclear fuel cycle is handled and stored in cylinders which are designed, manufactured, and maintained in accordance with the ASME Boiler and Pressure Vessel Code for unfired pressure vessels (Section VIII). There are presently more than 40,000 of these cylinders within the DOE Oak Ridge Operations complex currently used for the storage of isotopically depleted material (process tailings).

*Based on work performed at Oak Ridge National Laboratory and Oak Ridge Gaseous Diffusion Plant operated for the U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

These tails cylinders, in 10 and 14 ton sizes, are 48 in. in diameter, and are constructed of mild steel. Most cylinder storage facilities are open areas adjacent to the enrichment plants, where the cylinders are exposed to weather. Mild steel will corrode under these conditions, presenting the need for the monitoring of cylinders in storage to provide assurance that they have not deteriorated to the extent that they no longer meet wall thickness requirements for shipping and handling.

Cylinders used for transport of isotopically enriched materials are of heavier construction (5/8 in. wall construction rather than 5/16 in.) than these tails cylinders; they also see more activity than tails storage cylinders, and are therefore subject to relatively frequent visual observation and cleaning, as well as periodic pressure testing during which potentially hazardous conditions may be detected and evaluated. Tails cylinders in storage are subjected routinely only to periodic inventory; no cylinder inspection is required until the cylinders are either emptied or transported.¹

At one time, feed materials were transported exclusively in cylinders rated for 200 psi working pressure, the same design utilized for product (enriched) material. In frequent use in the fuel cycle, they too were subjected to periodic cleaning, inspection, and hydrostatic testing. Present practice, however, allows one-time use of a new 100 psi cylinder for UF_6 transport, after which it is used for tails storage at its destination. As a storage cylinder, it sees no further formal inspection. The cylinder yards are therefore occupied by three populations of storage cylinders:

1. Obsolete feed cylinders rated at 200 psi working pressure with nominal 5/8 in. wall thickness.
2. Thin (5/16 in.) wall cylinders code-rated for 100 psi working pressure, designed and built as storage cylinders.
3. Thin wall cylinders built as one-time use feed cylinders, then diverted to tails storage at the receiving site. These cylinders were built to the same specifications as the storage cylinders in category 2.

Categories 2 and 3, the thin (5/16 in.) wall cylinders, are the initial focus of this corrosion work.

The mild steels used in UF₆ cylinder construction are pressure vessel grades of plate steel, covered originally by ASTM A285, then since 1978 by ASTM A516 standards. They undergo a general atmospheric corrosion attack; they are also subject to pitting or other localized damage under exposure conditions where moisture is maintained in contact with cylinder surfaces, and particularly where this moisture may be contaminated by chemical species which dissociate in solution to form high-conductivity electrolytes. These corrosion processes will eventually reduce wall thickness to the point where the cylinder contents cannot be safely removed by liquefaction, but require sublimation emptying, a process both slow and costly. A cylinder with reduced wall strength due to corrosion would be more susceptible to handling damage during transport; damage to a cylinder during transport also involves potential environmental shock.

An extensive cylinder life study has been under way at the Paducah Gaseous Diffusion Plant to estimate the anticipated lifetime of storage cylinders.² Their data indicate that cylinders are generally expected to retain ASME Code qualification for at least another twenty years. The Oak Ridge work described in this paper supplements the Paducah study by examining conditions specific to the Oak Ridge cylinder yards, and determining local differences in corrosion rate related to cylinder yard location and storage condition of individual cylinders.

A four-faceted program is under way in Oak Ridge. Initial objectives are to determine the current condition of cylinders in outside storage at ORGDP, and mark those found to show marked deterioration. The program includes measuring corrosion rates to be anticipated in storage yards through preparation, deployment, and analysis of ASTM corrosion test coupons and commercial corrosion probes. It establishes and maintains a continuing inspection program of cylinders in storage, with statistically controlled measurements of wall thickness and records of corrosion activity. It further provides documentation through a complete data base to include cylinder manufacturing data, service history, corrosion observations, and location. Our work could be readily extended to any facility where cylinders are kept in storage for significant periods of time.

OAK RIDGE CYLINDER YARD CONDITIONS

Cylinder degradation is expected to take place primarily from exposure to the environment. Atmospheric corrosion of mild steel varies from 1 to 10 mils per year; UF₆ corrosion at the inside wall surface is expected to be less than 0.1 mil per year.^{3,4,5}

Weathering attack depends on the presence of surface moisture; corrosion rate is, therefore, influenced markedly by relative humidity. Surface temperature will affect both the corrosion kinetics and the presence of surface moisture. Position of the cylinder yards with respect to plant emissions and plant operating parameters will influence attack rate. Air circulation and sunlight will

also affect steel deterioration. Retention of moisture in contact with the steel surface, particularly where the moisture contains mineral salts, can promote localized attack or pitting corrosion where penetration rates can exceed general corrosion rates by a factor of 10 or more.

Oak Ridge rainfall varies in pH from about 3.7 to 5.2. The acidity of the rainfall has a highly localized character, even within an individual shower; however, not much rain is expected to fall outside the limits where significant acceleration of corrosion will occur (Fig. 1). Pitting due to particulate deposition is of more concern; scanning electron microscopy and energy-dispersive X-ray analysis have identified sulfur-bearing particles which have been deposited on the cylinder surfaces (Fig. 2).

Relative humidity in Oak Ridge is high; in the summer exposed steel surfaces at ambient temperatures can be expected to be covered with a thin film of condensed moisture at least half the time. Lower cylinder walls where the metal is in contact with solid UF₆ retain this moisture film over longer periods of time, and would be expected to suffer more corrosion damage.

Approximately 3000 storage cylinders in the K-1066-G yard were moved to a new location, K-1066-K in 1983-1984 (Fig. 3). Detailed observations were made on many of the cylinders at that time^{6,7}, revealing accelerated general attack and pitting in support areas shielded by wooden saddles on which the cylinders rested, and similarly accelerated corrosion in the head area of the skirted cylinders where the skirt accumulates moisture, rust, and dirt. Both of these areas showed pit depths of 60 mils, superimposed on more general wall thinning that, in total, reduced wall thickness in several cases to below the minimum value required by ASME Code criteria for working pressure of 100 psi.

General wall thinning on the upper surfaces of those cylinders was estimated at the time of the move to be only about 35 mils, even though wind rose patterns measured at ORGDP (Fig. 3) indicate that for a significant fraction of time cylinders located in the K-1066-G yard were downwind from the nearby steam plant. This amount of thinning is consistent with Paducah results,² which estimate less than 2 mils per year general attack rate.

Oak Ridge cylinder yard inspections are focussed on areas which represent potential problem regions. Visual inspection of the storage cylinders is under way. Ultrasonic measurement equipment has been adapted for easy use in the yards and field metallography techniques are being used to (a) determine the present status of stored cylinders, then (b) track the rate of deterioration. Cylinders identified as no longer conforming to code will be set aside for special handling. The wall thickness data, and any other pertinent observations (valve and plug conditions, damage) made during the projected periodic inspections, are being included in a computer data base.

A significant amount of data to be used as a guide for cylinder inspections is being obtained from the destructive evaluation (Fig. 4) of a Type P cylinder procured in 1951 from the Dallas Tank Company and fabricated of A285 steel. This

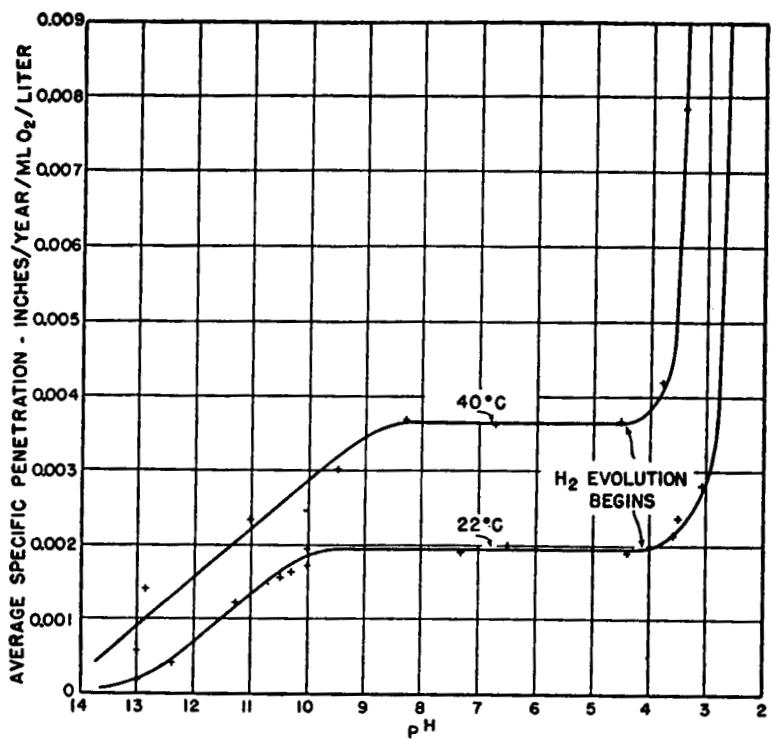


Fig. 1. Effect of pH on corrosion of mild steel (from Larrabee, C. P., "Atmospheric Corrosion of Iron," in *The Corrosion Handbook*, Uhlig, H. M., ed., John Wiley and Sons, Inc., New York, 1948). The pH of Oak Ridge rainfall varies from about 3.7 to 5.2, placing Oak Ridge corrosion rates in the plateau region of the curve.

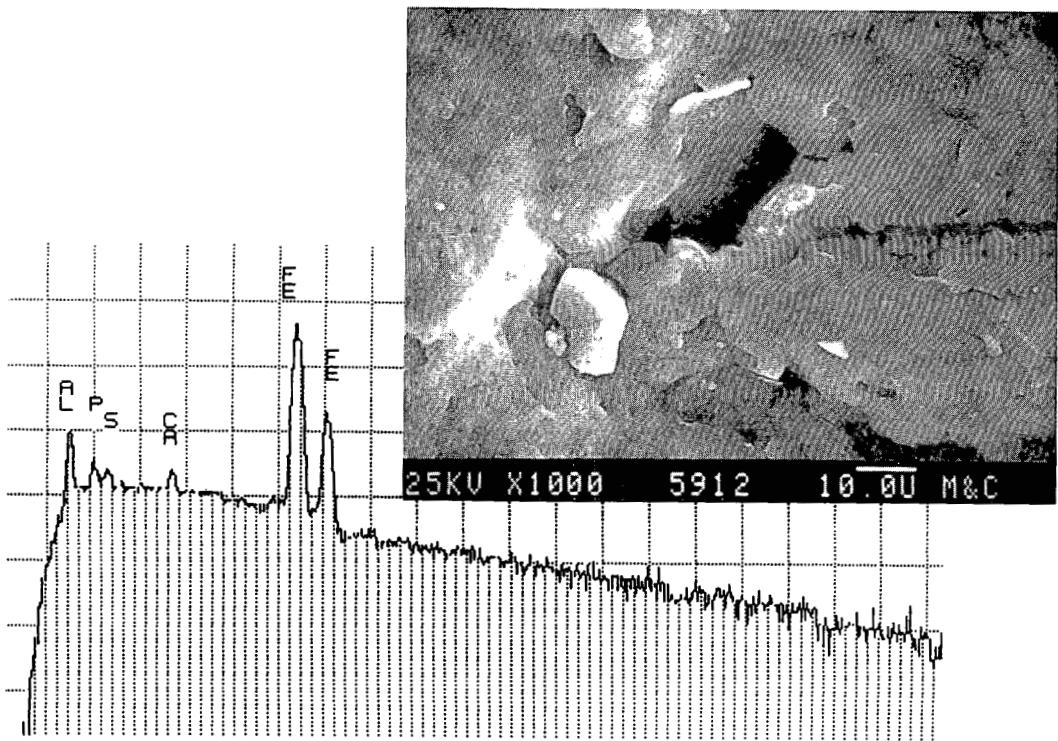


Fig. 2. Scanning electron microscopy and energy-dispersive X-ray analysis of tails cylinder surface particle. This particle contains sulfur; such particulates can initiate pitting corrosion.

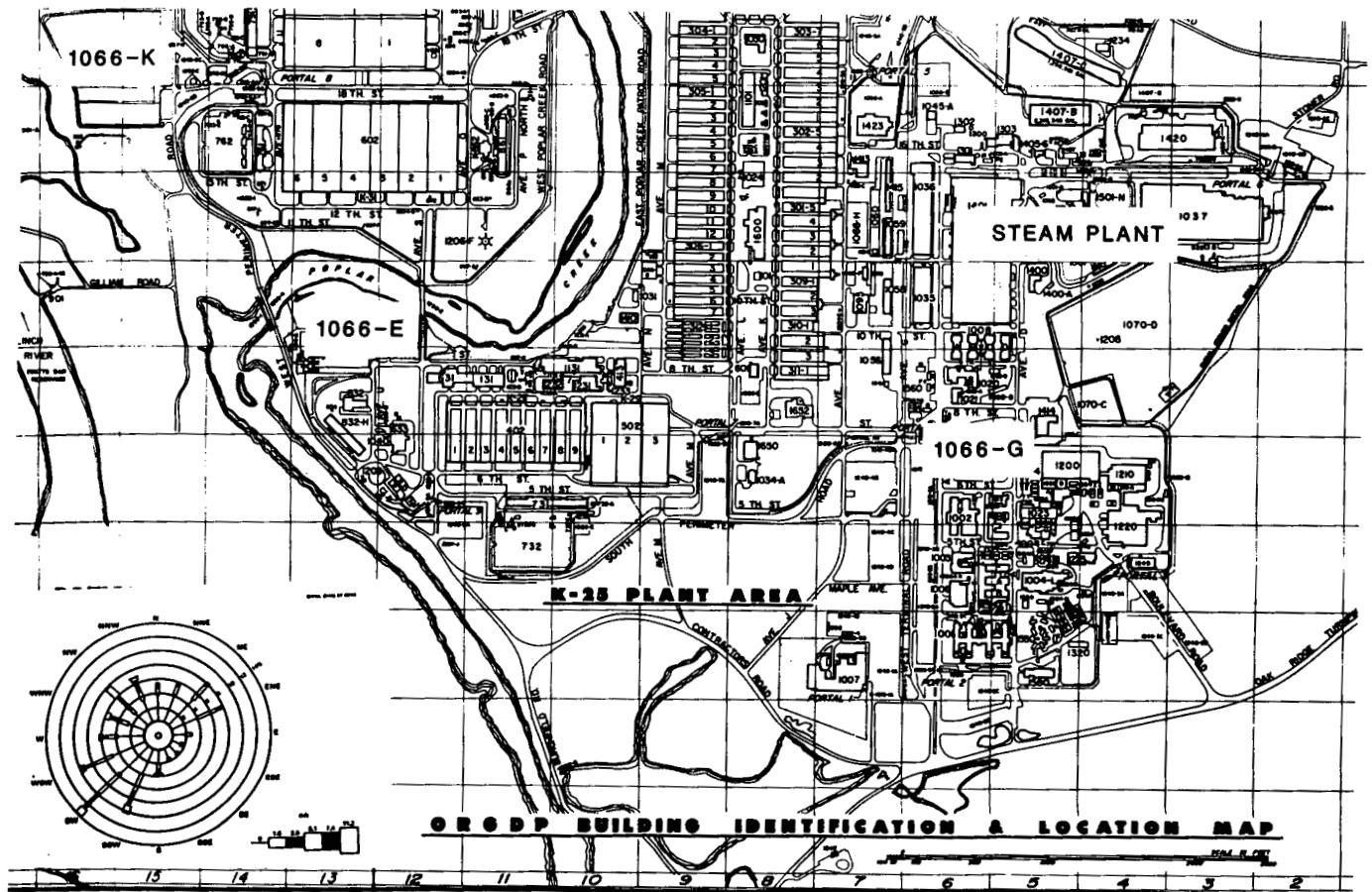


Fig. 3. Map of the Oak Ridge Gaseous Diffusion Plant, and wind rose data for the area. Approximately 3000 old tails cylinders were moved from the K-1066-G yard to the K-1066-K yard in 1983. Cylinders stored in the K-1066-E yard were fabricated after 1975. The most prominent wind in the area is from the southwest; however, night winds (second most prominent) are primarily from the northeast. The old K-1066-G cylinder yard was generally downwind from the steam plant during the evening hours.

cylinder was in continuous service from 1952 to 1985, when it was hydrostatically taken to failure as part of a cylinder rupture testing program.⁸ Ultrasonic measurements taken before cylinder sectioning showed significant wall thinning in the lower region of the head area, as well as near the plug weld. The cylinder was then sectioned; examination of the head wall confirmed this thinning (Fig. 5). Figure 5 also compares the general appearance of the inner and outer wall surfaces, confirming that the major corrosion processes are exterior. Field metallography and ultrasonic measurements obtained from this cylinder before sectioning are being compared to laboratory characterization of the sectioned cylinder. Similar data will be obtained from a Type 48Y cylinder, bought in 1980 from Modern Welding Company, and fabricated of A516 steel. These comparisons should confirm the validity of the field measurements to be taken in the cylinder yards. The sectioned cylinders will then be cut up to provide coupons for placement in the yards at ORGDP.

Information to date has identified areas such as (a) the bottom portions of the cylinders, (b) areas where the cylinders may have been in contact with the ground or with each other, (c) crevices where moisture and particulates may have been trapped, and (d) weld regions for special attention. Some such areas are shown in Fig. 6.

MEASUREMENT OF CORROSION RATES

Oak Ridge work to determine corrosion rates will center on the accurate measurement of environmental degradation of cylinder steel through the placement of steel coupons in the Oak Ridge cylinder yards. Testing will be conducted according to ASTM standards.

Three sets of steel coupons were chosen for study. Cylinders 287 and 9873 are being sectioned to provide actual cylinder material for evaluation. A285 steel is known to vary markedly in corrosion characteristics; therefore, samples of A36 steel plate will be included to define "worst case"

conditions. Two groups of each steel will be positioned in racks constructed in accordance with ASTM G50-76, *Standard Practice for Conducting Atmospheric Corrosion Tests on Metals*. A rack will be oriented in the cylinder yard facing south in accordance with ASTM G50-76. Other coupons will be placed in the yards to supplement the ASTM data with data more specifically reflecting corrosion rates in the regions of the cylinders where accelerated attack might be expected.

Each set of steel coupons will consist of several groups. A standard group of 4 in. * 6 in. (preferred ASTM G50-76) with no coupon surface preparation other than cleaning will be initially weighed and surface area measured in accordance with the provisions of ASTM G1-81. Data will be recorded in accordance with ASTM G33-72. All coupons will be removed at 1 year, 2 years, 4 years, 8 years, and 16 years. Weight gain will be recorded, then weight loss after scale removal.

A second group of coupons will be instrumented for time of wetness measurements. A third group of 1 in. coupons will be prepared metallographically before placement in the yards; these coupons will be sacrificed at each periodic removal for surface and microstructural characterization of the corrosion process as it proceeds from a surface representative of the bulk material. A fourth group will consist of 1 in. coupons taken from areas which include weld and heat-affected metal. These will also be sacrificed for surface and microstructural characterization. The 1 in. coupons will provide supplementary weight gain measurements.

Representative coupons from each group will be characterized as fabricated using Auger and X-ray photoelectron spectroscopy; optical, scanning electron and transmission electron microscopy; X-ray diffraction; and Raman and Mossbauer techniques as appropriate.

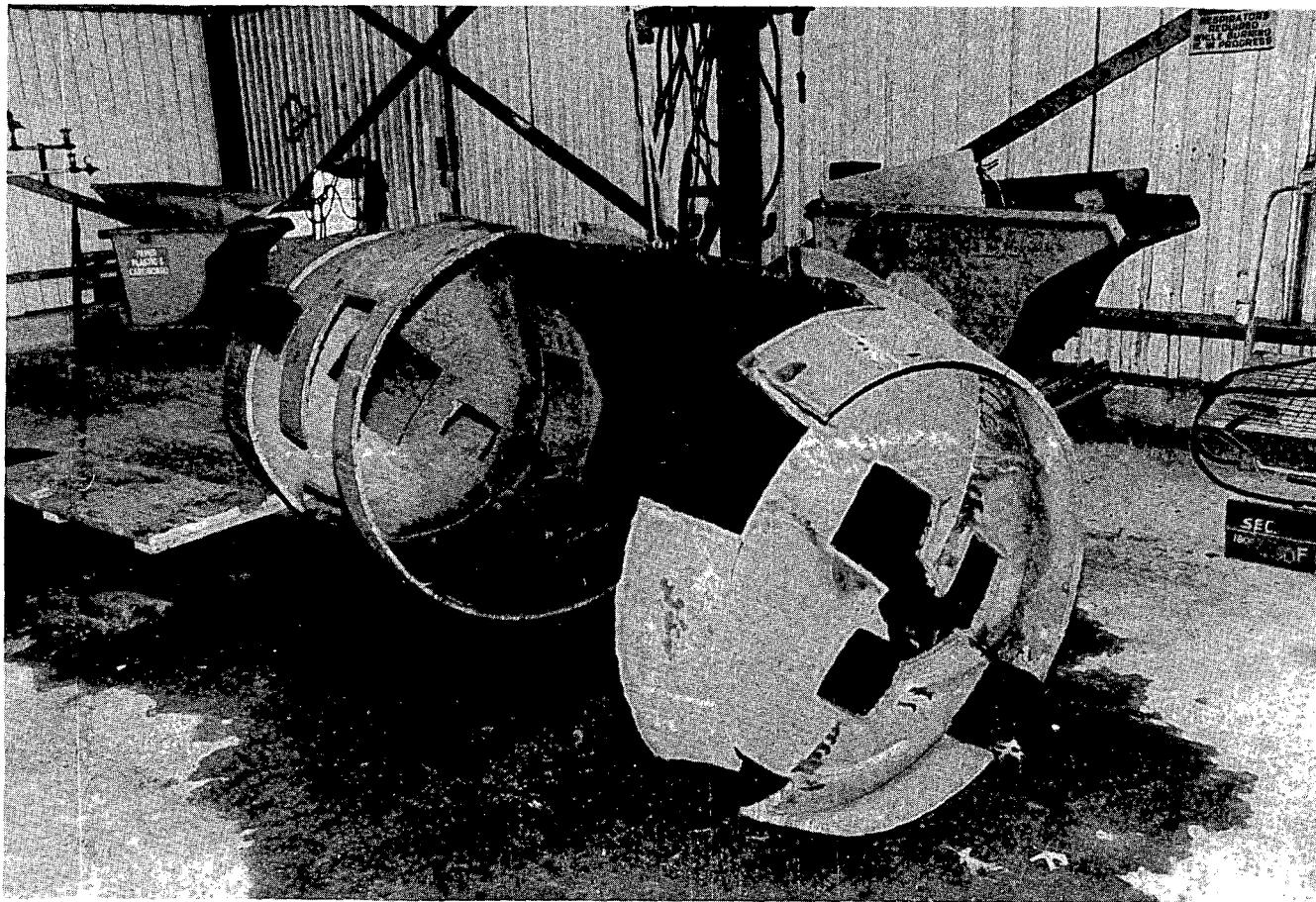
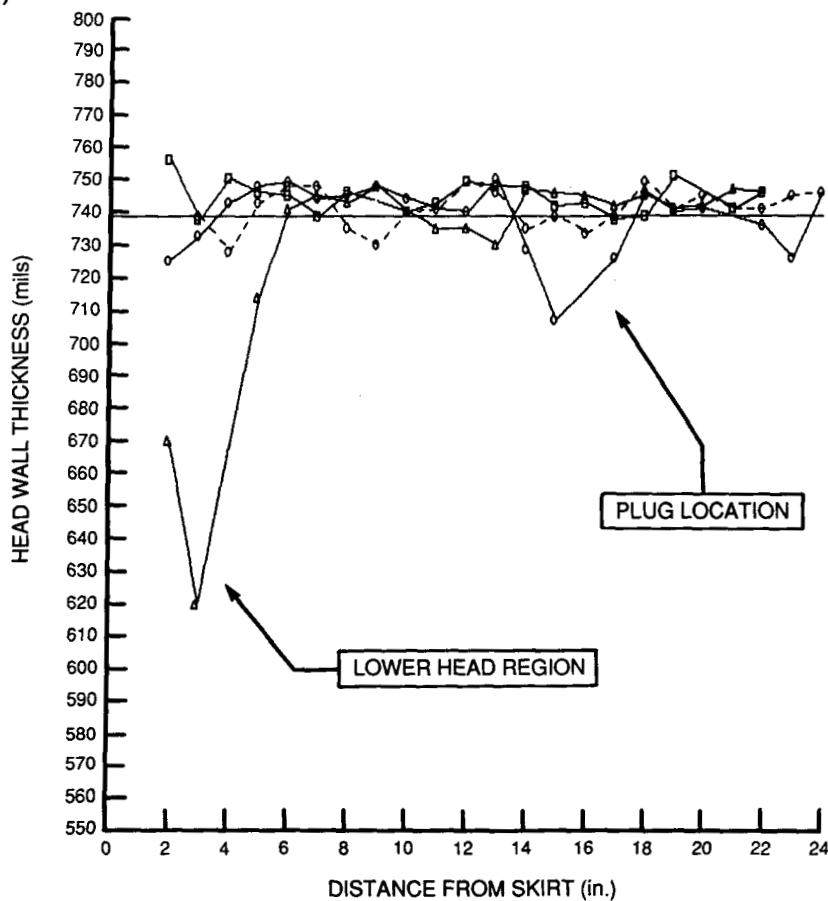


Fig. 4. Sectioning of cylinder 287. Field measurements were made on regions of this cylinder before sectioning; these will be confirmed through laboratory examination of samples cut from the same regions. Coupons cut from this cylinder will be placed in the cylinder yards.

a)

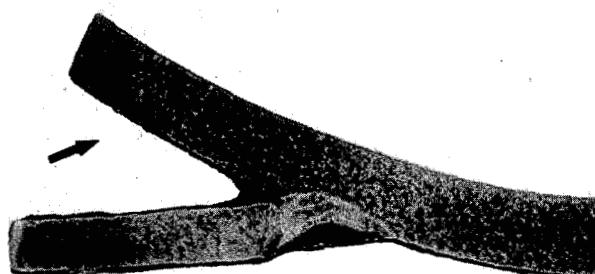
HEAD THICKNESS - PLUG END - # 287



b)

HEAD INTERIOR

WEATHERED SURFACE



SKIRT

Fig. 5. (a) Field ultrasonic measurements taken across head region of cylinder 287. Considerable thinning is observed in the lower head region and near the plug weld. (b) Section cut from the lower head region of cylinder 287. Corrosion is proceeding primarily from the outside; the interior surface appears smooth and undamaged. The thinning seen in Fig. 5(a) is confirmed after sectioning; optical thickness measurements in this region range from 0.75 to 0.66 in. Original thickness of this cylinder head was approximately 3/4 in.

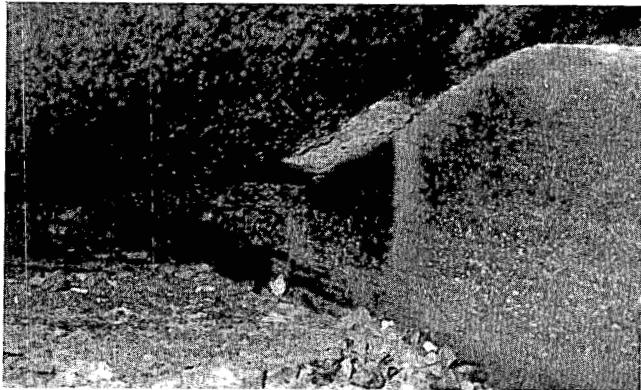
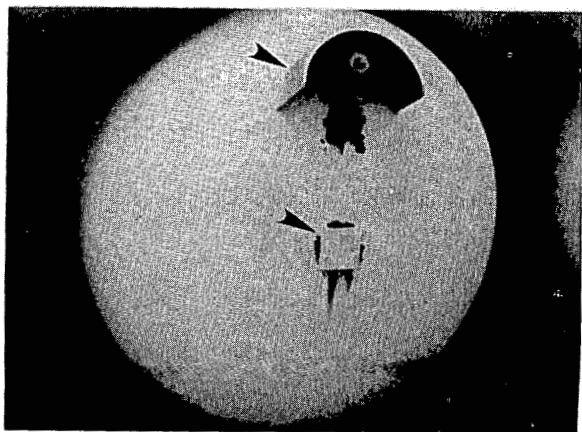
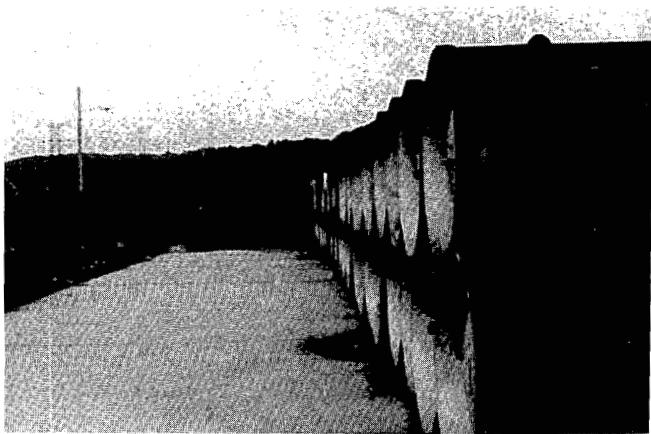


Fig. 6. Cylinders in storage at K-1066-K cylinder yard. Regions where accelerated corrosion attack might be anticipated are (a) lower head region, (b) areas in which dissimilar metals are in contact, (c) weld regions, and (d) areas where the cylinders rest on saddles. These regions are indicated with arrows.

CYLINDER YARD INSPECTIONS

Periodic visual inspections of the cylinder yards are to be supplemented by field metallography and ultrasonic testing on a statistically determined sampling of the cylinders. The field testing will emphasize those regions of the cylinders that have been identified for special attention.

Commercial E/R corrosion probes fabricated from A516 cylinder steel will be placed in the cylinder yards, and corrosion rate measured frequently. These commercial probes will give prompt notification of any change in environmental conditions leading to a major alteration in corrosion rate.

Inspection data will be entered into the data base described below.

DOCUMENTATION

A data base has been established following the guidelines set forth in *ANSI/ASME NQA-1 Basic Requirement No. 17 - Quality Assurance Records*. Data is entered in commercial dBase III spreadsheet format; and the data base is to be maintained using IBM-compatible personal computers. Cylinder manufacturing data, service history, and inspection records are included; records are flagged for cylinders identified through either inspection or history as requiring special handling.

SUMMARY

Tails cylinders in outside storage at ORGDP are corroding at rates which give a finite storage life as ASME code-qualified pressure vessels. General corrosion rates are estimated at less than 2 mils per year yielding a service lifetime anticipated at more than 50 years; however, cylinder areas where accelerated attack might be expected have been identified through (a) cylinder yard history and (b) examination of cylinders which have seen extended service, for special attention.

Field ultrasonic and metallography techniques have been developed for cylinder examination; periodic inspection (based on statistical sampling) of the cylinder yards at ORGDP is planned, using these techniques. A corrosion monitoring program for the Oak Ridge yards, based on ASTM procedures and using environmental corrosion test coupons, is under way. Documentation of cylinder condition will be provided through an IBM PC-based records system, using commercial software, and conforming to ANSI/ASME NQA-1 standards. The program is intended to assure that corrosion of these cylinders at ORGDP never reaches the point where the cylinders cannot be safely transported or pressurized for transfer.

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OVERVIEW OF THE FIVE-INCH PRODUCT CYLINDER

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ABSTRACT

Model 5A and 5B cylinders are used for transportation and storage of very highly enriched UF₆ product (VHE). The cylinders are manufactured from 5-inch, schedule 40 pipe with welded pipe cap cylinder heads. The Model 5B cylinder design has only recently been approved and has been included in DRO-651, revision 5, and the 1987 revision of ANSI Standard N14.1. The basic difference between the two models is that the Model 5A is fabricated from Monel, while the 5B is fabricated from nickel. A potential materials problem was identified with the Model 5A cylinder and resulted in a modification to the existing Model 5A cylinders. The operating history, including the cause of the change in the Model 5A design, results of recent proof testing, and the current cylinder specifications are discussed.

INTRODUCTION

The five-inch diameter product cylinder was designed to address nuclear criticality considerations for UF₆ enriched to greater than 30% U-235, as well as to assure materials compatibility with UF₆. The nominal five-inch diameter cylinder is an "always safe" geometry, and in use a minimum spacing between cylinders is maintained (24 inches center-to-center) to further ensure nuclear criticality safety. The five-inch product cylinder was designed to meet or exceed the design requirements of the ASME Boiler and Pressure Vessel Code, Division I, Section VIII pertaining to pressure vessels even though the

cylinder did not fall under the jurisdiction of the code. The physical design standards for the cylinder are given in Table 1.

TABLE 1 STANDARDS FOR THE FIVE-INCH CYLINDER

Diameter	5 Inches (12.7 cm)
Overall Length	35.6 Inches (90.5 cm)
Vessel Length	29.5 Inches (74.9 cm)
Wall Thickness	0.25 Inch (0.6 cm)
Minimum Volume	0.284 Cubic Feet (8.04 l)
Design Pressure	
Internal	200 PSI (1.38 MPa) at 250°F
External	22 PSI (0.15 MPa) at 250°F
	15 PSI (0.10 MPa) at -120°F
Hydrostatic Test Pressure	400 PSI (2.76 MPa)

The cylinders receive a hydrostatic pressure test at the manufacturing facility, and are also rehydro tested every five years or upon emptying if used for storage longer than five years. Three cylinder designs -- designated Model 5A, Model 5A Modified, and Model 5B -- have been utilized since the 1950s. The difference between these designs is the materials of construction which are given in Table 2.

The Model 5A cylinder is constructed of Monel 400 (70% Ni - 30% Cu); similarly, the Model 5A Modified is Monel 400 except the valve couplings or valve bosses are nickel. The Model 5B is constructed of nickel with Monel 400 trim (handles, foot ring, neck ring, etc.).

TABLE 2 MATERIALS OF CONSTRUCTION FOR THE FIVE-INCH CYLINDERS

Description	Model 5A	Model 5A Modified	Model 5B
Shell	Ni-Cu, ASTM B-165	Ni-Cu, ASTM B-165	Ni, ASTM B-161
Heads	Ni-Cu, ASTM B-165	Ni-Cu, ASTM B-165	Ni, ASTM B-161
Couplings	Ni-Cu, ASTM B-165	Ni, ASTM B-161	Ni, ASTM B-161
Neck Ring	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165
Foot Ring	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165
Dip Pipe	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165
Support Ring	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165	NI-Cu, ASTM B-165
Handles	NI-Cu, ASTM B-164	NI-Cu, ASTM B-165	NI-Cu, ASTM B-164
Valve Protector	Carbon Steel	Carbon Steel	Carbon Steel

MODEL 5A

The Model 5A cylinder was designed and procured in the early 1950s for the handling of highly enriched UF_6 . The material of construction (Monel 400) was selected for its excellent corrosion resistance to fluorinating materials as well as for its resistance to atmospheric corrosion. This corrosion resistance provided a container which would not consume the product or result in external corrosion thereby altering the tare weight and/or reducing container integrity. Two problems have been identified with the Model 5A cylinders. The first problem was encountered in the mid 1960s when a customer was using empty cylinders as cold traps for niobium pentafluoride, a use outside the design service conditions. During this service the Monel 400 cylinder developed stress corrosion cracking. The customer was informed of the damage to the cylinder, and the cold trapping of this compound was discontinued.

The second problem was more serious and resulted in a modification to the Model 5A cylinder. In the early 1970s, routine inspection of the cylinders revealed valve coupling (boss) cracking. This cracking was investigated and determined to have been caused by liquid metal penetration of the grain boundaries, resulting in embrittlement and grain boundary cracking. The embrittling agent was the silver solder used in the brazing of the cylinder valve into the cylinder. The investigation also indicated that the embrittlement and cracking occurred during valve replacement and not during initial valve installation. During valve replacement the boss area is heated to the silver solder liquidus temperature, the old valve removed, the area cooled, the boss inspected, and a new valve inserted and brazed in place. The heating of the boss to the brazing temperature permitted residual silver solder from previous valve installations to penetrate the Monel 400 grain boundaries which is a cracking mechanism for this material. This problem resulted in a change in boss material from the original Monel 400 to nickel as explained below.

MODEL 5A MODIFIED

To accomplish the boss material changeout in the 5A cylinders, a program was instituted in the 1970s to systematically replace the valve bosses at the time of cylinder maintenance or during routine hydrostatic retesting. The resulting cylinders were termed Modified Model 5A or Model 5A Modified. To date, all Model 5A cylinders in the normal product flow cycle have been converted to the Model 5A Modified specification.

The cylinder modification was designed to meet or exceed the requirements of the ASME Boiler and Pressure Vessel Code, Version I, Section VIII (the

"Code"). However, at the time of the modifications the Portsmouth facility did not hold a "Code Stamp," and therefore the modified Model 5A cylinders were not coded pressure vessels. This fact had very little impact upon the use of the cylinders, and the cylinders were subsequently listed in the two governing documents for UF_6 cylinders: ORO 651, "Uranium Hexafluoride-Handling Procedures and Container Descriptions," and ANSI N14.1, "American National Standard for Packaging of Uranium Hexafluoride for Transport." However, recently drafted regulations from both national and international agencies governing the shipping of UF_6 have brought under scrutiny the voided "Code Stamp" on the Model 5A Modified cylinders. An extensive investigation into the fitness for service of the modified cylinder was therefore undertaken.

The investigation focused on demonstrating that the cylinder modifications met the intent of the code and that cylinder integrity had not been compromised. An engineering study encompassing design calculations, review of modification procedures, and proof testing (hydrostatic pressure testing) of three cylinders was performed to verify the integrity of the Model 5A Modified cylinders. The study concluded that the modification to the Model 5A cylinder did not affect cylinder integrity based on its demonstrated reliability in the field for up to twelve years, acceptance of the modification procedures by the "National Board of Boiler and Pressure Vessel Inspectors," and the proof testing results. The hydrostatic pressure test results for three modified cylinders were compared to a result of a similar test performed in the 1960s prior to the modification. The rupture pressures were considered similar for all four tests and are shown in Table 3. Failure mode and location were also similar.

In late 1981 and early 1982 five of the Model 5A Modified cylinders were found to have surface cracks on the cylinder head (near or adjacent to the head-to-boss weld) and also apparent weld interface cracks. The weld failure mode was lack of fusion on the valve boss-to-cylinder head weld caused, in part, by the use of a nickel welding electrode instead of the specified Monel electrode. The cylinder head cracking failure mode was similar to that of the Model 5A valve boss cracking noted in the early 1970s. In modifying the Model 5A cylinders, the silver solder from the brazing of the valve had run down the side of the nickel valve boss and penetrated the Monel 400 cylinder heads. The silver solder then embrittled the Monel 400 which resulted in the cracking of the cylinder heads. In an effort to eliminate the silver solder embrittlement failure mode, the Model 5B cylinder was designed.

TABLE 3 PROOF TEST DATA FOR 5A MODIFIED CYLINDERS

Cylinder Type	Modification Date	Wall Thickness	Rupture Pressure	% Volume Increase
5A	NA	$\frac{1}{4}$ " (0.6 cm)	8250 psi (56.7 MPa)	--
5A Modified	1986	$\frac{1}{4}$ " (0.6 cm)	7950 psi (54.8 MPa)	21
5A Modified	1979	$\frac{1}{4}$ " (0.6 cm)	7200 psi (49.6 MPa)	21
5A Modified	1977	$\frac{1}{4}$ " (0.6 cm)	8600 psi (59.3 MPa)	23

MODEL 5B

The design of the Model 5B UF₆ cylinder was completed in 1983 and presented to the Martin Marietta Energy Systems, Inc., three-plant, UF₆ Cylinder and Cylinder Valve Specification Committee. The cylinder is identical to the Model 5A and 5A Modified cylinders in dimensions (Ref. Table 1). The only difference between the two cylinder designs is that the Model 5B shell and head are constructed of nickel instead of Monel. This cylinder is subject to the same testing and quality assurance measures as the other five-inch cylinder designs; it has been procured as a "Coded" pressure vessel and is being phased into routine use for handling enriched UF₆. The Model 5B design has been included in ORO 651, Revision 5 and presented to the ANSI N14.1 Committee for incorporation into the 1987 Standards edition.

CURRENT STATUS

The five-inch product cylinders in use at this time are restricted to the Model 5A Modified and

Model 5B designs. Almost all of the original Model 5A cylinders have been reworked. The few Model 5A cylinders in use are being used for extended storage and have not been emptied since the modification program was initiated. Of the five-inch cylinders presently in service, 148 are of the Model 5B design which were procured in 1983-1984. Currently, 250 Model 5B cylinders are on order, with another 250 to be procured in 1989 at which time all off-site product shipments will be made using Model 5B cylinders. The remaining Model 5A Modified cylinders will then be limited to on-site UF₆ transfer and storage. To date, the Model 5B cylinder has been in service for four to five years, and, based on this limited operating experience, has demonstrated satisfactory operating performance. A fabrication procedure used to initially install the valves in some of these cylinders has been tentatively identified (at the time of the writing of this paper) as causing boss cracking during revalving of these cylinders. The results of this investigation will be discussed when the paper is presented.

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MAXIMUM CYLINDER FILL LIMIT EVALUATION

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ABSTRACT

The maximum fill limit for the variety of cylinders used by DOE and the remainder of the nuclear industry is a key element in maintaining a safe and efficient operation. The fill limits are based on several important parameters. These factors are discussed in the report along with the rationale utilized and the conservatism provided.

INTRODUCTION

The nuclear industry utilizes a wide variety of cylinders designed specifically for the safe handling and transport of UF_6 . The cylinders are manufactured to strict engineering specifications which are developed with close regard to the properties of UF_6 . The cylinders range from the very small laboratory container designed for handling gram quantities of UF_6 to the large 48-inch diameter containers for use in shipping and handling up to 14 standard tons of UF_6 . Each cylinder and each cylinder design have unique characteristics and anomalies which limit the maximum quantity of UF_6 to be withdrawn into the cylinder. The maximum quantity of UF_6 which can be safely contained in a cylinder is based on several parameters and was developed to insure safe handling during typical in-plant activities as well as routine transportation requirements. The paper which follows examines each of the parameters, exhibits the conservatism provided within each of these parameters, and shows how the parameters are used in determining maximum fill limits. Additionally, recent modifications to fill limits will be examined along with the rationale for the changes.

DEVELOPMENT OF CYLINDER FILL LIMITS

The requirement for establishing maximum fill limits for UF_6 cylinders is obvious; personnel safety. The characteristics of UF_6 , especially

density and coefficient of expansion, in addition to the hazards must be recognized. Safety must be attendant in all phases of the operation whether it be feeding, filling, or shipping. Safety is the key directive which governs both design and operation. A flaw in a cylinder design, an overfilled cylinder, an overheated cylinder, or a cylinder containing excess volatile compounds can all lead to disaster, an occurrence not desired with the current sensitive world environment. The procedures and practices for safe handling of UF_6 have evolved by the interaction of many companies and countries. Consistency within most countries exists, however, commonality between countries frequently surfaces differences in rationale used to develop the guidelines. The IAEA is pursuing the possibility of common international policies. Shipper and receiver rationale must be equitable to allow a continuation of the international nuclear industry.

The primary and most widely used documentation for maximum cylinder fill limits, in addition to many other related topics, has been ORO-651, "Uranium Hexafluoride: Handling Procedures and Container Descriptions," and ANSI N14.1, "Uranium Hexafluoride-Packaging for Transport." The Department of Energy (DOE) document ORO-651 has been compiled and issued based on the extensive experience at the DOE facilities. The latest revision of this document was issued during September 1987. Although the original intent of the document was for U. S. Government use, the audience has been expanded to an international scope.

The ANSI N14.1 document has also been recently revised (October 1987) to include several changes in industry practices. Initially issued in 1971 as the private industry version of ORO-651, this document was developed with international participation around the framework provided by ORO-651. Both documents are currently viewed as complements. The tables, in fact, in both ORO-651 and ANSI N14.1 regarding maximum fill limits are virtually identical. Therefore, the basis for international consistency of maximum fill limits has been established. But what parameters are evaluated to determine the maximum amount of UF_6 which can safely be withdrawn into a certain design or size cylinder? Each of these parameters will be explored in detail in the next section.

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PARAMETERS AFFECTING FILL LIMITS

There are four parameters, in addition to standard UF_6 characteristics, which govern the maximum cylinder fill limit. These are 1) cylinder volume, 2) maximum cylinder design temperature, 3) purity of the UF_6 and 4) safety factor or "Free Volume" desired in the containment. These items are not listed in priority order since all are integral and essential ingredients in the determination process. By discussing each of the factors independently, then collectively, the conservatism designed into the fill limits will become very apparent.

The initial factor to be discussed is the volume of the cylinder. Cylinder design and procurement specifications for all cylinders purchased for DOE facilities require, among other requirements, a minimum internal volume. The actual volume, which is greater than the minimum in virtually every case, is certified by evaluating the capacity of the cylinder by filling the vessel with water, weighing, and correcting to the standard temperature of 60 degrees Fahrenheit. The DOE facilities have employed, over the years, the use of both certified and non-certified cylinders. At the present time, only certified cylinders are used in shipments from DOE facilities and the use of cylinders containing non-certified volumes is strictly minimized.

The comparison of the actual cylinder volumes to the minimum specifications is appropriate to determine the average "additional" volume available for UF_6 residence. Two widely used cylinders were chosen as candidates to exhibit the comparison; the Model 48X 10-ton cylinder and the Model 48G 14-ton cylinder. The evaluation was conducted at the Paducah Gaseous Diffusion Plant by collecting data in a random fashion from cylinders in service. The model 48X cylinders were constructed in the early 1950s and are currently used to transport enriched product between facilities. The minimum specification cylinder volume is 108.9 cubic feet. The sampling of 83 cylinders revealed an average volume of 112.06 cubic feet or the existence of more than 3.0 cubic feet (2.9 percent) additional volume over that used in maximum fill limit calculations.

The model 48X cylinders were all constructed to the identical specification and showed the expected cylinder-to-cylinder variation. The Model 48G cylinders, as a further example, were constructed to similar specifications and by numerous fabricators. Three separate samplings were conducted at Paducah involving 53 cylinders. The average certified volume in each of the three samplings was 143.70, 143.86, and 144.06 cubic feet. While the averages are remarkably similar, the lowest value found in any of the 53 cylinders was 139.82 cubic feet, still well above the minimum of 139.0 cubic feet.

The conclusion to be reached from this single parameter is that significant conservatism relative to safe fill limits exists in cylinder volume alone. Why not use the actual volume for determining maximum fill limits? The minimum volume is standard for each cylinder type, provides a consistent limit for each type and use, and minimizes errors that could exist if each

individual cylinder had a unique maximum limit.

The next parameter which has an effect on the maximum limit is the maximum cylinder design temperature. While the term implies a mechanical or structural limit, the maximum actually refers to the operating temperature of the UF_6 . The term maximum design is to be considered synonymous with maximum operating for the balance of the report. The coefficient of expansion of UF_6 between the solid and liquid phases is extremely high and an operating temperature above the maximum allowable could cause the expanding liquid to completely fill the internal volume of the cylinder and create hydraulic forces. The forces could exceed the structural yield value for the cylinders if the temperature was allowed to increase further. A violent explosion could result from an uncontrolled heating of a cylinder containing UF_6 .

The maximum design temperature for the heavy-wall cylinders is typically 250 degrees Fahrenheit and the maximum design temperature for most of the light-wall cylinders is 235 degrees Fahrenheit. It must be mentioned at this point that some of the light-wall cylinders constructed during the 1950s and early 1960s were catalogued with a maximum cylinder design temperature of 300 degrees Fahrenheit although this temperature was never approached at a DOE facility.

The percent of the available volume to be occupied when a specific quantity of solid UF_6 is heated to a liquid in a closed cylinder is a direct function of temperature. If the temperature is increased, the liquid expands further and occupies an increased volume. If the temperature is reduced, the converse is true. The maximum cylinder fill limits are constructed by using these maximum temperatures. In actual operating practice at virtually all DOE sites where solid UF_6 is vaporized and/or liquefied, the highest temperature reached is substantially below this maximum. Once again, this provides an additional measure of conservatism. The typical temperature will range from a low value of just over 200 degrees Fahrenheit to about 225 degrees Fahrenheit. These lower temperatures will reduce the volume the liquefied UF_6 will occupy by approximately three percent.

A quantification of the above scenario can be constructed by using a cylinder containing 25,000 lbs. of UF_6 . The UF_6 , when heated to 250 degrees Fahrenheit, will occupy additional cubic feet when compared to the same quantity heated to only 225 degrees Fahrenheit. The reduction in the temperature (maximum versus typical operating) consists of a conservatism factor of over three percent. Why not use the typical operating temperature? While DOE sites maintain a lower temperature, other UF_6 handling facilities routinely approach 235 degrees Fahrenheit to attain required flow rates. Therefore, it is considered prudent to establish a consensus value, in this case the maximum design temperature, on which to base maximum fill limits. Additional safety can be obtained by lowering operating temperatures. The maximum design temperature and the certified water weight capacity are stamped on each cylinder nameplate.

Various chemicals are present in UF_6 as impurities. The concentration of these low molecular weight compounds has been explored theoretically and relationships established. These low molecular weight compounds are more volatile than UF_6 and can be found in both the liquid and the vapor phase as a cylinder is being heated. The impurities in the UF_6 after withdrawal from the gaseous diffusion process are typically found to be air components, oxygen and nitrogen, refrigerant, various fluorides and HF. The isotopic enrichment process will cause the lighter weight gases to move toward the product withdrawal segment of the plant. A portion of these materials will be withdrawn into product cylinders. The tails withdrawal stream, containing the greater portion of the heavier depleted uranium isotopes, will be void of any significant concentrations of the impurities.

The maximum fill limits documented in both ORO-651 and ANSI N14.1 are based on a minimum purity factor of 99.5 weight percent UF_6 . This provides for a maximum impurity of less than one-half percent in all cascade material streams, including tails and product. The DOE diffusion plants conduct routine uranium, isotopic and radiochemical measurements on these streams to determine the quality of the material, including purity. The results of the evaluation procedure for FY 1987 are summarized as follows:

Stream Analyzed	Number of Samples	Results (wt.% UF_6)	
		Average	Range
Product	31	99.973	99.95-100.0
Tails	26	99.976	99.95-100.0

The calculations determining the maximum fill limits for UF_6 cylinders have assumed the presence of up to one-half percent impurities and have required the availability of ample void space for residence of these gases during the heating cycle. The general assumption has been to consider no solubility of the gases in UF_6 . In actuality, as a cylinder is being heated for vaporization or liquefaction, the impurities contained within the void space above the solid UF_6 will be compressed only a slight amount. The compressibility factor and solubility of the gases has been established through testing and analytical evaluation. The further heating of the UF_6 will cause an increase in the rate at which the light gases condense and reenter solution with UF_6 .

The FY 1987 data in the table above is typical for material withdrawn from the Paducah cascade. A review of previous years will provide similar results and verify that the purity of UF_6 product or tails from the diffusion plants is extremely high. Although the fill limit calculation allows for a maximum of one-half percent impurities, the actual percent of impurities present in cylinders is, under the most pessimistic conditions, only ten percent of this value. Therefore, the void space available for light gases or impurities has been overstated, yielding conservatism and additional space for UF_6 expansion.

The final parameter which has a significant impact on the magnitude of UF_6 which can be safely placed in a cylinder is the safety factor or "free volume." This factor has been discussed to a degree in the purity section and is defined as a specific percentage of the minimum internal volume of the cylinder. The volume is provided to allow residence space for light gases and/or additional expansion room for UF_6 , if needed and is the only one of the parameters which is operator variable.

The safety factor was originally defined as five percent of the minimum internal volume of the cylinder at the maximum design temperature of 250 degrees Fahrenheit for heavy-wall cylinders. The factor was reduced to three percent at the 235 degree Fahrenheit level for the light-wall cylinders to be used for long-term storage of tails material. This distinction recognizes the slight difference in purity when comparing product and tails. Recent changes in the fill limit rationale have increased the safety factor to five percent for the light-wall cylinders. The light-wall cylinders are typically used for transporting and storage of depleted material, which is inherently purer, and require less "free volume." The increase in the safety factor simply increases the confidence in the use of the cylinders, but also allows the overall fill limit rationale to be more consistent: five percent safety factor (based on the minimum cylinder volume) at the maximum design temperature of the cylinder.

A review of the four key parameters affecting the determination of maximum cylinder fill limits shows a consistent rationale which has been adopted internationally. Each of the factors has been determined in such a manner to yield a minimum of a five-percent margin of safety. This provides limits which are moderately "forgiving" in daily operating scenarios; but are based on factual evidence.

COMPARISON OF LIQUID AND SOLID FILL LIMITS

The fill limit evaluation is designed to be of assistance either while a cylinder is being filled, fed, or transferred. But how does this rationale apply when the UF_6 has solidified and the cylinder has been prepared for shipment?

A cylinder containing liquid UF_6 should be handled only the minimum amount necessary to place the vessel into appropriate saddles for solidification. Liquid cylinders are not transported on standard conveyances over the road; only cylinders containing solid UF_6 . The elevated risk and danger is present while the UF_6 is in liquid form. Therefore, the maximum fill limits, as well as other restrictions, should be developed around this segment of the operation. Then, as the material solidifies, the shipper and receiver can be well assured of a cylinder which is safe under routine operating and shipping conditions.

The only credible scenario in which a cylinder containing UF_6 would be a hazard during transport is to involve the cylinder in a major fire during an accident. The cylinder would have to be engulfed in the uncontrolled heat source for a significant period of time. There have been no accidents involving material in transit from a DOE facility in which a fire has engulfed the cylinder

causing a hydrostatic rupture. There have been no actual scenarios anywhere in the world in which this has happened. Therefore, no special precautions need be applied for transportation of UF₆ although some organizations are considering the use of overpacks for routine shipments of all material for safety purposes in addition to criticality. A conservative maximum fill limit developed with regard to liquid UF₆ will provide an equally conservative condition for a cylinder containing solid UF₆. Until April 1986, there had never been a limit expressed on the percent of the cylinder volume which could be occupied by solid UF₆. At that time, a Notice for Proposed Rulemaking was issued by the U.S. Department of Transportation which would limit the occupied volume of solid UF₆ at 68 degrees Fahrenheit to 61 percent. The value was developed by converting the maximum fill limit for Models 48X and 48Y, as presented in ORO-651, to a percent of internal volume by use of the density of UF₆ at 68 degrees Fahrenheit (317.8 lb/ft³). The calculated volume of UF₆ was then compared to the minimum acceptable volume for the cylinders to yield a value of approximately 61 percent. By strict application of the DOT regulation, the majority of DOE light-wall cylinders filled with tails material have been rendered non-shippable without the initiation of drastic action. This action would be to heat cylinders and vaporize or liquid transfer a portion of the material from the cylinders. The risks associated with this action far outweigh the shipping of the cylinders containing solid UF₆ occupying greater than 61 percent.

How do the parameters previously mentioned really translate into shipping limits? The application of the parameters discussed earlier allow for reasonable and safe shipping limits. The two primary limits are as follows:

- Cylinders with a maximum design temperature of 250 degrees Fahrenheit

- Require five percent safety volume

- Density of UF₆:

$$\begin{aligned} @ 250 \text{ degrees Fahrenheit} &= 203.3 \text{ lb/ft}^3 \\ @ 68 \text{ degrees Fahrenheit} &= 317.8 \text{ lb/ft}^3 \end{aligned}$$

- Occupied volume =

$$\frac{203.3}{317.8} \times .95 = 60.77\% \text{ volume}$$

This calculation yields a volume very similar to the value determined by DOT, although with a more firm analytical basis.

- Cylinders with a maximum design temperature of 235 degrees Fahrenheit

- Require five percent safety volume

- Density of UF₆:

$$\begin{aligned} @ 235 \text{ degrees Fahrenheit} &= 207.1 \text{ lb/ft}^3 \\ @ 68 \text{ degrees Fahrenheit} &= 317.8 \text{ lb/ft}^3 \end{aligned}$$

- Occupied volume =

$$\frac{207.1}{317.8} \times .95 = 61.91\% \text{ volume}$$

While the second calculation exceeds the DOT regulatory limit, it is based on realistic parameters not included by DOT. In an effort to resolve the issue, DOE has petitioned DOT through priority and emergency exemptions to accept a 62 percent value for the lower temperature (light-wall) cylinders.

RECENT NATIONAL AND INTERNATIONAL ACTION ON MAXIMUM FILL LIMITS

Several recent modifications have been made to cylinder fill and shipping limits. These changes are documented in both ORO-651 and ANSI N14.1 - 1987.

The ORO-651 guideline listed on the "UF₆ Cylinder Data Summary" states:

For DOE gaseous diffusion plant tails with UF₆ purity in excess of 99.5% the shipping limit is 28,000 lbs. for cylinders with 8,880 lbs. water capacity or greater (142.35 cubic feet).

In like manner, ANSI N14.1 - 1987 states:

Fill limits (for cylinder Models 48T, 480, 480M, 48H, 48HX, and 48G) are based on 235 degrees Fahrenheit maximum UF₆ temperature and minimum UF₆ purity of 99.5 percent. The allowable fill limit for tails UF₆ with a minimum purity of 99.5 percent may be higher but shall still not result in a cylinder ullage of less than five percent when heated to the cylinder design temperature of 235 degrees Fahrenheit based on actual certified volume.

Each of the statements is equivalent and supports the position established in this paper. The excerpts from both ORO-651 and ANSI N14.1-1987 are based on conditions surrounding the filling of a cylinder with liquid UF₆; however, as stated previously, the resulting volume occupied by the UF₆ after solidification will also be acceptable.

CONCLUSION

The development of maximum fill limits for UF₆ cylinders has been completed and documented both nationally and internationally. The limits have been modified recently to include new information pertaining to certain cylinder types. The evaluations have recognized the key parameters which have an effect on the validity of the limits, while establishing a firm foundation in safety. The primary factors include minimum/actual internal cylinder volume, maximum/actual operating temperatures, high purity of the diffusion plant UF₆, and the maintaining of an adequate safety or "free" volume. By utilizing these factors, maximum fill limits can be constructed for consistency and safety. The limits were developed around the filling of cylinders with liquid UF₆ and have resulted in realistic limits based on consistent rationale. A very conservative maximum fill limit for liquid UF₆ conditions will result in equally conservative volume occupancy limits for solid UF₆.

OVERPACKS AND PROTECTIVE PACKAGING FOR 30-INCH UF₆ CYLINDERS

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ABSTRACT

The development and use of protective packaging often referred to as overpacks for transport of 30-inch diameter uranium hexafluoride cylinders is not new. The generation of 21PF-1 and 21PF-2 protective shipping packages developed under Department of Transportation (DOT) rules for a specification package are currently still in use. However, problems developed with the 21PF's in 1982 which led to a DOT proposed rulemaking in August 1984. Action to complete the rulemaking has not been completed. However, a recently published American National Standards Institute standard on Uranium Hexafluoride for Transport (1987) has placed greater emphasis on protective packaging quality assurance and periodic inspection. The chemical hazard of uranium hexafluoride has also received recent attention by the International Atomic Energy Agency in their 1987 report on "Recommendations for Providing Protection During the Transport of Uranium Hexafluoride".

Nuclear Packaging, Inc. (NuPac) recognized a need for an advanced state of the art protective package and designed, tested, obtained an U.S. Nuclear Regulatory Commission certificate of Compliance and a DOT certificate of competent authority for IAEA regulations. The designation of this package is the UX-30.

This paper will discuss the UX-30 design considerations, testing approach used, and test results obtained which supported approval of the Certificate of Compliance by NRC.

Finally, a recommendation is made to complete the DOE proposed rulemaking and incorporate the revised ANSI N14.1 Standard-1987.

I. INTRODUCTION

The topic of this paper is "Overpacks and Protective Packaging for 30-Inch Uranium Hexafluoride Cylinders". The terms overpacks and protective packaging are synonymous. It is the outer packaging used to enclose cylinders containing enriched uranium hexafluoride (UF₆) exceeding 1 weight percent U²³⁵. A US Department of Transportation Specification 21PF-1 protective overpack (reference 1, 49 CFR 178.121) has been used in the transport of isotopically enriched UF₆ since about 1968, during which time thousands of overpacks have been manufactured and placed into service. Photos of these overpacks in service in Japan, Europe and the United States are presented.

Approximately 85-90% of the 21PF-1s are controlled by Japan and Europe, with the USA controlling the balance, or 10-15% (200 to 300 containers). Interestingly enough, the Eastern block countries do not use overpacks. Soviet enriched UF₆ is sent to Riga by rail then loaded into overpacks for shipment elsewhere in Europe and to the USA.

There have been two foreign and two US manufacturers of 21PF-1 overpacks: one in Germany, one in Italy and two US manufacturers. There is one active 21PF-1 manufacturer in the US today. UF₆ moves in worldwide commerce: Sweden, Japan, France, United Kingdom, Germany, and USA all have conversion plants to convert UF₆ to uranium oxide and reactor fuel.

Three incidents in 1982 required radiological response forces to respond to apparent problems caused by the inleakage of water to the 21PF-1 overpacks. Several 21PF-1s were rejected for water leakage, rotted wood in the step joint, holes in the outer shell, and rusting of the outer shell and structural members. Proper maintenance of the 21PF-1 is required. Lack of maintenance, design flaws and poor manufacturing practices created potential safety problems - such as listed in Table 1.

Table 1

Current Safety Problems 21PF-1

Water Absorption (200 pounds plus)
Moderator
Degraded mechanical properties
Steam explosion in case of fire
Wet-freeze-thaw cycle effects
Corrosion
Degraded mechanical properties
Thermal resistance in case of breached shell
Incomplete Foaming
Thermal resistance
Energy adsorption
Not Made to Specification
Italian soft wood (end-glued small pieces) frame
Not Stamped with Tare Weight
Cannot determine water content

The recognition of these potential safety issues led the DOT to publish a proposed rulemaking on August 10, 1984 (reference 2), relative to repair of existing 21PF-1s and the design modification of new 21PF-1s. Meanwhile, General Electric and Westinghouse, both involved in UF₆ conversion to UO₂ for commercial reactor fuel, obtained fabricators to build a stainless steel version of the 21PF-1 design, but with one important design change. The joint between the two hemispherical cylinders was inverted to prevent water inleakage. These 21PF-1 containers were licensed by the NRC under Certificate of Compliance 4909, July 1983. At the same time, Nuclear Packaging, Inc. began development of an alternative packaging design which ultimately came out under NRC C of C 9196, December 1984. The list of NRC licensed overpacks is summarized in Table 2.

Table 2

NRC Licensed Overpacks

GE-21PF-1 General Electric	US NRC C of C 4909, Rev. 8 expires 2-28-93. IAEA C of C, Rev. 5 Expires 11-30-88. This is a stainless steel version of the 21PF-1. Fabricated by Getchell & Sons, Southfield, RI.
W-21PF-1 Westinghouse	US NRC C of C 4909, Rev. 8 Expires 2-28-93. IAEA C of C, Rev 5 expires 11-30-88. This is a stainless steel version of the 21PF-1. Fabricated by Precision Metals, Wilmington, NC.
UX-30 Nuclear Packaging	US NRC C of C 9196, Rev. 1 Expires 12-31-89, IAEA C of C, Rev 1 expires 12-31-89. Deploys polyurethane foam and has been drop tested to demonstrate meeting IAEA Safety Series 6, 1973 requirements. Fabricated by NuPac subcontractors under NuPac QA program.

II. CHANGING UF₆ SHIPPING REGULATIONS

One person died as a result of the large release of liquid and gaseous uranium hexafluoride in a 1986 production plant accident in Oklahoma, United States of America. The International Atomic Energy Agency (IAEA) has taken the lead to assess the adequacy of UF₆ transport regulations considering both the radiological and chemical hazards posed by this material. The IAEA's "Recommendations For Providing Protection During the Transport of Uranium Hexafluoride" IAEA-TECDOC-423 June 1987 (Reference 3), report states that depleted, natural or low-enriched UF₆ has a subsidiary risk classification of 8 (corrosive) and Special Provision 174 also applies which states (in part) "...packaging should conform at least with the requirements of American National Standard ANSI N14.1 - 1982..." (Reference 4). The ANSI N14.1-1982 Standard has been recently revised and published as ANSI N14.1-1987 (Reference 5). The 1987 Standard addresses two areas of concern: (1) Quality Assurance and (2) Outer Protective Packaging.

The essential QA effect is to require a licensee-user to have a documented quality assurance program, meeting subpart H, title 10, CFR, Part 71 or ANSI/ASME NQA-1 for those quality-related activities associated with procurement, maintenance, repair and use of the cylinder and the protective packaging.

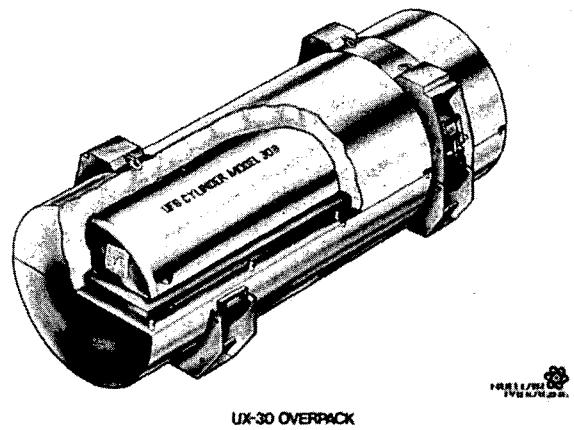
The Outer Protective Packaging Section requires in-service inspections, maintenance, periodic inspections, tests and recertification, and repairs all in accordance with a qualified quality assurance program and by properly qualified welders.

III. DEVELOPMENT OF AN ADVANCED DESIGN PROTECTIVE PACKAGE (OVERPACK)

As a result of the 21PF-1 problems identified in 1982, Nuclear Packaging undertook the development of a newly designed and licensed overpack for the 30-inch UF₆ cylinders. In the summer of 1983, NuPac had completed the design, overpack prototype fabrication, full scale prototype drop tests, and the draft SARP. This status report and the licensing, strategy, design features, operational features, and hypothetical accident behavior were discussed at a non-public DOE meeting in Oak Ridge on August 11, 1983.

The main points of this status report will be reiterated briefly for this paper. First, the Licensing Strategy was to incorporate a handling assembly that was not a structural part of the package, 10 CFR 71.31(c) and (d), and thus not a part of the NRC licensed package design and to draw upon recent licensing experience and understanding of the NRC's regulatory concerns.

In this artist's conception (Figure 1), the primary design features of the UX-30 are shown.



UX-30 OVERPACK

Figure 1. UX-30 Overpack

Note that the geometry is a clean, uncluttered right circular cylinder. The dimensions are:

Exterior 43.5" dia. x 96" long
Interior 31.0" dia. x 82.75" long

The stainless steel inner and outer shells form the basic sidewall structure. The six inch closed cell polyurethane foam provide thermal insulation and impact protection. The closure consists of a full clam shell opening for the 30-B cylinders. There is a deeply steeped lid lip with a weather seal and finally a ball-lock sheer pin fastening.

The overall nominal shipping weights are overpack 2130 lbs., handling assembly 450 lbs. (varies with alternative designs) 30-B cylinder 6420 lbs. for a total of 9000 lbs. for each package. Five packages can easily be hauled on a truck/trailer to meet legal weight limits of 80,000 gross vehicle weight.

The operational features involve three main considerations: Use, maintenance and tiedown assembly interface. The UX-30 can be used easily by operations personnel in the field with ease and convenience. The quick opening ball lock flush pin closure allows the clam shells to be closed without the use of tools, bolts or auxiliary equipment. The UX-30 package, with its transport saddle, can be lifted and stacked with a minimum of work steps and time (Figure 2). For maintainability, the UX-30 has smooth, clean, even surfaces and all joints are seal welded for watertight integrity. The stainless steel construction is corrosion resistant and requires no painting. The seals are fully protected from handling damage. The foam has been tested to prove that it has no water absorption capability. The tiedown assembly interfaces well with the UX-30 package. The tiedown assembly can be customized to user needs without relicensing. It lifts the overpack and lid with a single setup. No tools are required to attach the four over-center adjustable latches. It requires no spreader bar, thus allowing for fork lift use.

Stacking features are flexible; for example, one overpack can be stacked on top of another overpack or a lid of an overpack can be placed on top of another overpack, etc. There is an optional tie-down using an existing bolt pattern or chain binders. Loaded UX-30s are carried five to a truck trailer (Figure 3). The hypothetical accident behavior has been analyzed. Two primary events are discussed in the SARP: impact resistance and fire resistance. The impact resistance features are the use of an integral strongback for the UF_6 cylinder protection. The use of the cylindrical geometry provides for maximum impact resistance and weight efficiency. The prototype UX-30 was dropped 30 feet to demonstrate successful impact resistance behavior. The integrated membrane design provides optimum puncture resistance. Ductile stainless steel shells guarantee no rupture and no tearing during puncture events. The foam is fire resistant as demonstrated by sample testing. The maximum UF_6 cylinder temperature following a 30 minute 1475°F fire is less than 150°F.



Figure 3. Loaded UX-30's

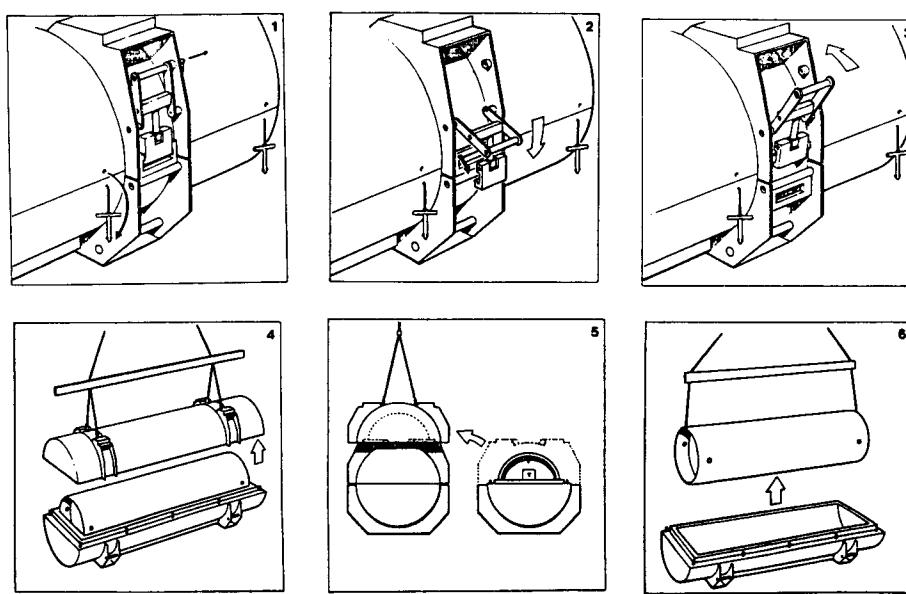


Figure 2. Handling Sequence

The testing sequence involved loading the prototype UX-30 with a 30-B cylinder which in turn has been filled with 5,023 lbs. of steel punchings to simulate the maximum payload of 5,020 lbs. of UF₆. The UX-30 was then lifted for a thirty foot drop onto a concrete pad. The corner drop deformed the outer steel shell and foam as predicted. A forty inch drop onto a six inch diameter puncture pin was then conducted in an orientation so that the point of impact was aligned with the valve shell deformation, but not penetration. The inner shell displayed only a scuff mark where the rim of the 30-B cylinder came in contact with the end surface. Thus, the drop tests proved that the UX-30 overpack would protect the 30-B cylinder in accordance with the prescribed NRC impact tests.

Application for the Certificate of Compliance for the UX-30 was made January 13, 1984. First round of NRC questions were received July 30, 1984. There were no significant technical concerns and the amendments were directed at loading and maintenance procedures to be referenced. On August 31, 1984, NuPac submitted Rev. 1 to the SARP which responded to these questions. An additional set of questions and revision to the SARP yielded satisfactory results. The NRC issued Rev. 0 for the UX-30 Certificate of Compliance USA/9196/AF on December 17, 1984. The Department of Transportation issued the Competent Authority Certification for a Fissile Radioactive Materials Package Design Certificate USA/9196/AF, Rev. 0 on July 15, 1985, which certified the package as meeting International Atomic Energy Agency Safety Series No. 6 Regulations. Subsequent minor revisions to the SARP and amendments to the NRC and IAEA C of Cs have been made which result in the current C of C expiration date of December 31, 1989, stated in NRC C of C, Rev. 1 and IAEA C of C, Rev. 1.

IV. DEPARTMENT OF TRANSPORTATION PROPOSED RULE-MAKING

The Department of Transportation, Materials Transportation Bureau and Special Programs Administration issued a notice of proposed rulemaking "Modifications to DOT Specification 21PF-1 Overpacks" on August 10, 1984. The purpose was to propose modifications to the 21PF-1 overpack to alleviate problems which have resulted from water inleakage, retention and subsequent outleakage. This refers back to the events discussed in the introduction to this paper which occurred in 1982. Comments have been made by interested parties, but to date there has been no disposition of this proposed rulemaking. There will be papers presented later in this session which will address the proposed design changes and adequacy of the 21PF-1 package. These changes are relatively significant in that they require changing the skin and stepped joint from carbon to stainless steel, providing a waterproof seal which will be sacrificed to prevent venting in a fire, improve gaskets, allow for material substitutions (wood type), require tare weighing and labeling, and implement inspection requirements prior to operation.

Nuclear Packaging, Inc. believes that there is a need for a realistic examination by a 21PF-1 user to determine if his package meets upgraded requirements and if not, what the economics are in retrofit or replacement. Retrofit of an existing

package or construction of a new 21PF-1 will result in a package which is permitted to be used outside of the USA only under the DOT "Special Arrangement Certificate". The use of a Nuclear Packaging, Inc. UX-30 is authorized by IAEA Certificate USA/9196/AF Rev. 1 which indicates that there is full compliance with IAEA Safety Series 6 1973 edition regulations.

Nuclear Packaging, Inc. believes that there may be an inherent fatal flaw in the use of phenolic foam in the 21PF-1 package. Phenolic foam with a fire retardant boric/oxalic acid per se is an acceptable material. However, there is no evidence that repeated freeze-thaw cycles on foam that has gotten wet, frozen, thawed, gotten somewhat dry, wet again, frozen, thawed, etc. will hold up. There has been work done, which ORNL will discuss today, that shows phenolic foam when either wet or frozen, will display compressive strength and fire resistance. But reported evidence that foam which had been repeatedly subjected to a wet-freeze-thaw cycle and which will still be able to support the impact and fire has not been reported. More conclusive results than have been reported in ORNL reports K/SS-471, K/D-5400 Rev. 3, K-2057 Rev. 1, K/PS-1128 and K/PS-5068 (Reference 6 through 10) have not been found.

Furthermore, the action of water (or sea water) on phenolic foam to produce chloride ions and hence stress corrosion cracking of stainless steel is possible. This would raise doubts about the ability of the stainless steel version of the 21PF-1 package to withstand chloride corrosion.

V. CONCLUSION

The problems exhibited by some 21PF-1 containers in 1982 have not been resolved. Although changes in the design of the 21PF-1 have been proposed, their formal adoption by US Department of Transportation has not been accomplished. Similarly, retrofits to the existing 21PF-1 casks have not been authorized. A conclusion to the proposed rulemaking needs to be accomplished.

It is recommended that DOT issue a final rulemaking which:

1. Recognizes existence of NRC certified overpacks which are commercially available,
2. Recognizes IAEA's concern for the chemical hazards of UF₆; and
3. Recognizes American National Standards Institute Standard ANSI N14.1 - 1987 recently published as "Uranium Hexafluoride Packaging for Transport" which tightens the manufacturing and routine inspection, repair and quality assurance requirements.

ACKNOWLEDGMENTS

The author wishes to acknowledge the contribution of Richard T. Haelsig who reviewed and provided constructive comments on this paper. Mr. Haelsig was principally involved in the testing and licensing of the UX-30 package in 1983-1984 in his

roles at Nuclear Packaging, Inc. cumulating as the President 1984-1987. The UX-30 design was created by Larry J. Hansen and John D. Kent also of Nuclear Packaging, Inc.

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SAFETY EVALUATION OF THE TRANSPORT CONTAINER FOR NATURAL URANIUM-HEXAFLUORIDE UNDER FIRE ACCIDENT

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1. ABSTRACT

In Japan, it is scheduled to start this year, the land transportation of 48Y-cylinder filled with natural uranium-hexafluoride (UF₆) from Tokyo Harbor to the enrichment plant at Ningyou-pass in Okayama Prefecture.

In this study, heat resistance capability analyses of 48Y-cylinder filled with 12.5-ton of natural UF₆ under the condition of 800 °C for the duration of 30 minutes were conducted.

We developed simple modeling technique for heat resistance analysis of UF₆ packages which gives conservative and reasonable solutions, with two-dimensional or axi-symmetric models. Also, the effect of heat protective covers was examined analytically. The evaluated items of this 48Y-cylinder are as follows:

- (1) The hydraulic breaking of cylinder by expansion of UF₆ in liquid phase.
- (2) The breaking of the cylinder by vapor pressure increasing of UF₆ and the effect of the strength lowering of highly heated steel.
- (3) Leakage of UF₆ by melting soft solder of the valve and loss of integrity of containment

According to the results of preliminary analyses, temperature of 48Y-cylinder without heat protective instruments reached the melting point of soft solder (about 203 °C) in 2 or 3 minutes after the analysis had started. From this point of view and public acceptance (PA), we determined to mount the heat protective covers, 14 mm thickness, in the both ends of the cylinder.

Fire resistance analyses of the 48Y-cylinder filled with UF₆ were carried out. As material properties of natural UF₆, the data which were obtained through

the study of PNC (Power Reactor and Nuclear Fuel Development Corporation)³⁾ and the values listed in other papers^{1),2)} were used. For numerical analysis, ABAQUS, a general purpose F.E.M. program for non-linear problems was used.

2. OBJECT FOR ANALYSIS

Figure 1 shows the 48Y-cylinder made of steel. In this analysis, the effects of heat protective covers and the valve protector (for drop impact) are taken into consideration.

2.1 VALVE PROTECTORS

Figure 2 shows the detail of the valve protector for drop impact. The valve protector is made of carbon steel.

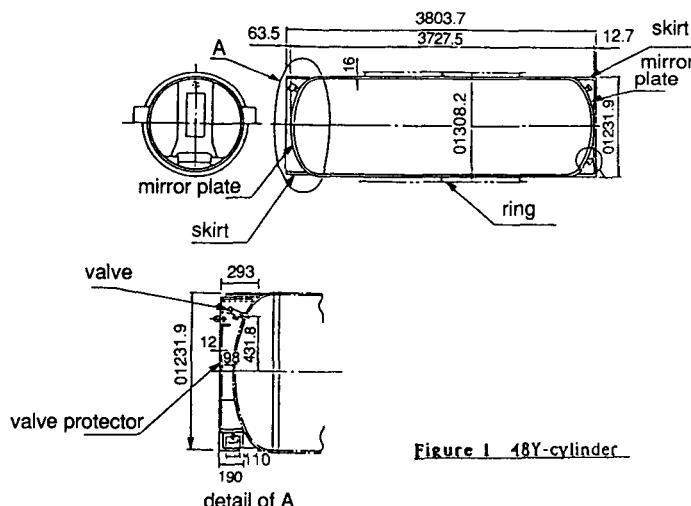


Figure 1 48Y-cylinder

Table 1 Material Properties of Mat

Name of Material	Glass Cloth	Ceramic-Fiber Cloth	Ceramic-Blanket
Critical Temperature	300 °C.	1000 °C.	1260 °C.
Thickness	0.5 mm	2.0 mm	9.0 mm
Weight	0.46 kgf/m ²	1.18 kgf/m ²	0.80 kgf/m ²
Tensile Strength	170 kgf/30mm	8.3 kgf/10mm	0.732 kgf/cm ²
Thermal Conductivity (at 800 °C)		0.18	0.16 (unit : kcal/m hr °C)

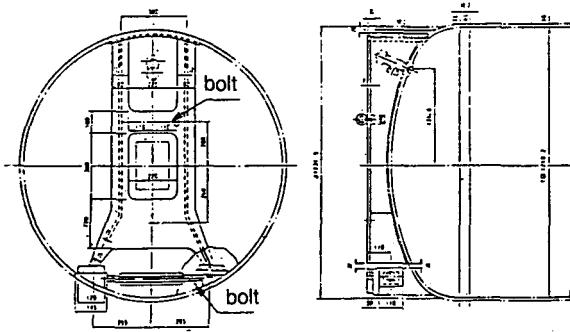


Figure 2 Detail of Valve Protector

2.2 HEAT PROTECTIVE COVER

Figure 3 shows the heat protective covers mounted to the 48Y-cylinder. As total weight of the container is limited by the domestic road traffic control law, the covers are mounted in only both ends of the cylinder. Figure 4 shows the detail of the laminated layer structure of the mat. Silicon is sprayed on the outer surface of the glass cloth for the purpose of absorption of humidity. Table 1 shows the material properties of the mat.

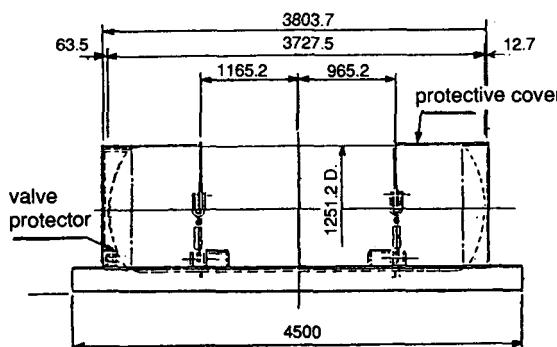


Figure 3 48Y-cylinder with Heat Protective Covers

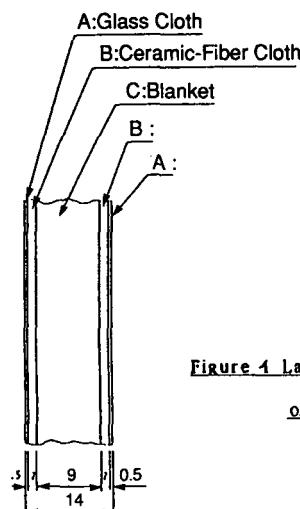


Figure 4 Laminated Structure
of Heat Protective Cover

3. NUMERICAL ANALYSIS OF 48Y-CYLINDER FILLED WITH NATURAL UF6

3.1 OUTLINE

Fire resistance analyses of the 48Y-cylinder filled with UF6 were carried out under the condition of 800 °C for duration of 30 minutes.

Two cases of analyses were conducted in the same way as the fire resistance tests. CASE-1 was performed by the cylinder without heat protective covers, and CASE-2 with the covers. In CASE-2, the effects of the valve protector for drop impact was considered. As material properties of UF6, the data which were obtained through the study of PNC was used³⁾.

3.2 EVALUATED ITEMS

In case that 48Y-cylinder filled with natural UF6 meets a fire accident, expected damages of the cylinder are predicted as follows:

- (a)The hydraulic breaking of cylinder by expansion of UF6 in liquid phase.
- (b)The breaking of the cylinder by vapor pressure increasing of UF6 and the effect of the strength lowering of highly heated steel.
- (c)Leakage of UF6 by melting soft solder of the valve

Evaluation methods for each item is as follows:

(1) HYDRAULIC BREAKING

As triple point of UF6 is about 64 °C, above this point, UF6 is in liquid phase and expands in accordance with temperature rise. Figure 5 shows the relation of temperature and density in liquid phase¹¹⁾. In this analysis, the volume of UF6 is computed from the temperature distribution of UF6 and temperature-density relation.

In case that the volume of UF6 (V_{UF}) reaches to be equal to the capacity of the cylinder (V_{CYL}), i.e.

$$V_{UF} = V_{CYL} \quad (1)$$

then, hydraulic breaking is considered to occur.

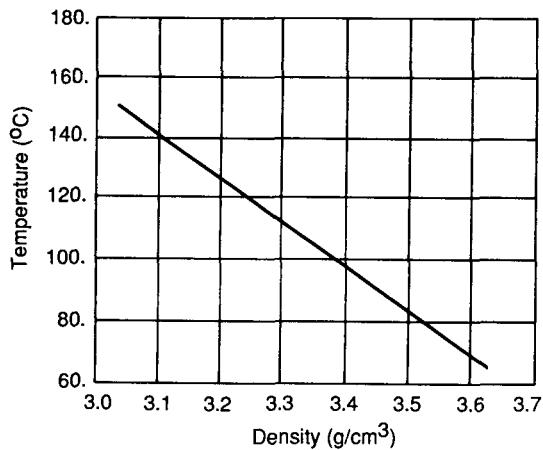


Figure 5 Relation of Temperature and Density of UF₆ in Liquid Phase

(2) BREAKING BY VAPOR PRESSURE

Figure 6 shows the curve which displays the relation between temperature and saturated vapor pressure¹⁾.

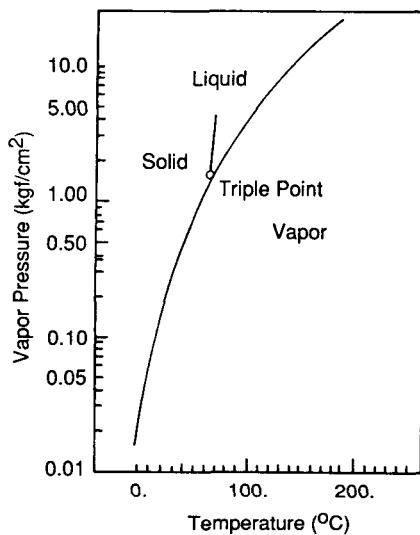


Figure 6 Relation of Temperature and Saturated Vapor Pressure of UF₆

Internal pressure of 48Y-cylinder (P_{in}) is calculated from the maximum temperature of UF₆ and the temperature-vapor pressure relation. P_{in} is estimated by the maximum temperature of UF₆.

From the maximum temperature of the cylinder, critical internal pressure of the cylinder (P_{cr}) is calculated using Tresca's condition.⁶⁾

Tresca's condition is as follows:

$$P_{cr} = \sigma_y \log \left(\frac{D_2}{D_1} \right) \quad (2)$$

σ_y ---- yield strength of steel
 D_2 ---- outer diameter of cylinder
 D_1 ---- inner diameter of cylinder

In this study, we modeled the 48Y-cylinder as thin shell structure and in case that all part of the cylinder is in plastic state, the cylinder is judged to be broken.

Figure 7 shows the relation between temperature and yield strength of steel, equivalent to SA-516 grade 60.⁶⁾ According to this relation, σ_y decreases with temperature rise.

And in case that P_{in} reaches to be equal to P_{cr} , i.e.

$$P_{in} = P_{cr} \quad (3)$$

then, the cylinder is judged to be broken by vapor pressure. At normal temperature, P_{in} is equal to 58.0 (kgf/cm²).

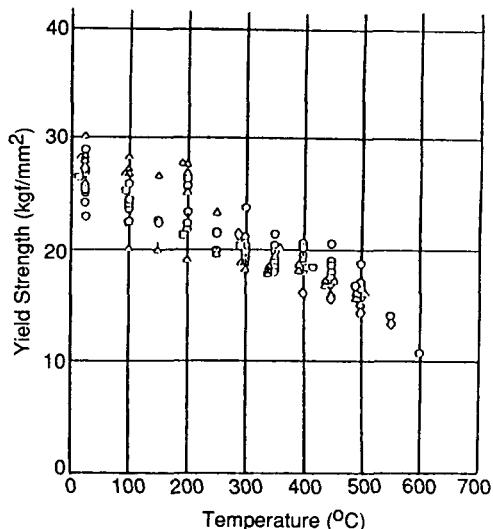


Figure 7 Relation of Temperature and Yield Strength of Carbon Steel

(3) LEAKAGE OF UF₆

The soft solder of the valve begins to melt above its melting point (203 °C). Then the cylinder loses its integrity of containment.

3.3 MODEL AND METHOD FOR NUMERICAL ANALYSIS

Behavior of UF₆ at high temperature is very complicated. To simulate all of the phenomena under the condition of fire accident, three-dimensional analyses are required. But three-dimensional analysis is very time-consuming and is not efficient.

Therefore in this analysis, we proposed to use two different axi-symmetric models and developed simple modeling technique which produces conservative and reasonable solutions.

As is mentioned in the previous section, the following values are required for safety evaluation of 48Y-cylinder under fire accident condition:

- (a) Maximum temperature of UF₆.
- (b) Maximum temperature of cylinder.
- (c) Temperature of the valve.

Figure 8 and 9 show the analysis models. Evaluating items of each model are listed as follows:

- model-A---temperature of UF₆
- model-B---temperature of cylinder and valve

Each of these two models gives a conservative solution for each evaluating items. Hence safety evaluation of 48Y-cylinder under the fire condition were conducted totally by combining the solutions of these two models in this analysis.

Features of each model can be expressed as follows:

(1) FEATURES OF model-A (Figure 8)

As UF₆ contacts with inner surface of cylinder overall in this axi-symmetric model, the area of contact surface between cylinder and UF₆ is about 1.2-1.5 times as large as that of actual cylinder. It means that a larger quantity of heat flux flow into UF₆. So this model could give conservative solutions with the regard to the temperature of UF₆. Because equivalent specific heat value is used, total heat capacity is equal to that of actual cylinder and UF₆.

(2) FEATURES OF model-B (Figure 9)

As mentioned before, model-A is conservative with regard to the temperature of UF₆, but not so with the regard to the temperature of the cylinder and valve.

In model-B, temperature of UF₆ is given as fixed boundary condition. Temperature of that boundary is set to the maximum temperature of UF₆ obtained from the results of model-A. And the surface level change of UF₆ in liquid phase has been neglected. Surface level is fixed at that in solid phase.

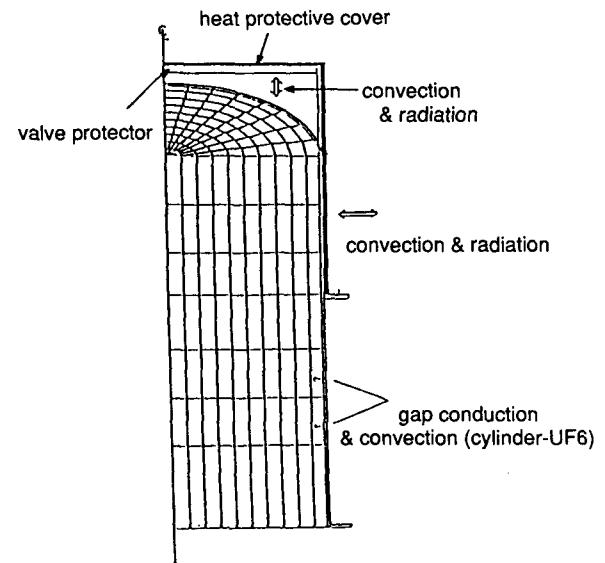


Figure 8 Features and Boundary Conditions of model-A

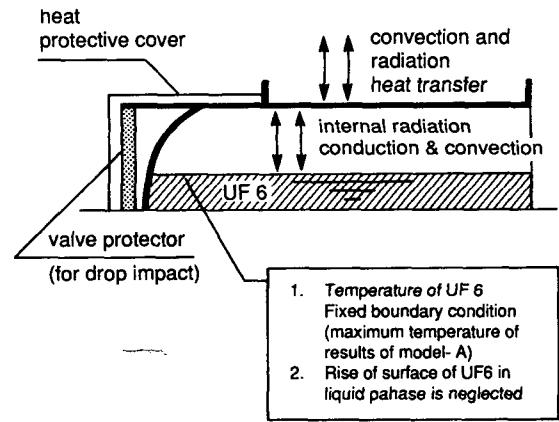


Figure 9 Features and Boundary Conditions of model-B

So total quantity of heat flux into UF₆ is less than the actual 48Y-cylinder and this model gives conservative estimation with regard to the temperature of the cylinder and valve.

3.4 BOUNDARY CONDITIONS FOR THE ANALYSES

Boundary conditions of the analyses are as follows:

- (1) Heat transfer between the environment and the container is caused by radiation and forced convection heat transfer. Emissivity of the outer surface of the cylinder is 0.60. Emissivity of the heat protective cover is 0.80. According to the IAEA regulations, emissivity of ambience was set to 0.90. Forced convection heat transfer coefficient was set to 3.60 (kcal/m² hr °C) by experimental formula of horizontal cylinder^{4),5)}.

(2) Heat transfer between the cylinder and UF₆ is caused by conduction (in solid state) and natural convection (in liquid state) heat transfer. Moreover effect of thermal gap is considered at the first stage of the fire accident. This thermal gap grows because of sublimation of UF₆. In this analysis, the effect of thermal gap is treated as gap conduction heat transfer. Value of gap conductance is according to the results of tests for material property of UF₆ conducted by PNC³⁾.

(3) The effect of the valve protector for drop impact was considered in CASE-2. As the structure of the valve protector is not axi-symmetric physically, equivalent value is used as the material property of the valve protector in this analysis. In CASE-2, heat transfer between the protector (and heat protective cover) and the cylinder is caused by radiation and conduction and natural convection heat transfer of the cavity air.

(4) Initial temperature is set to 38.0 °C according to IAEA regulations.

(5) Ambient temperature during fire accident is set for 800 °C. And after fire accident has terminated, it is 38.0 °C.

(6) Emissivity of free surface of UF₆ is set for 1.0.

3.5 MATERIAL PROPERTY

Table 2-4 show material properties. Properties of natural UF₆ is based on some papers^{1),2)} and experimental data of PNC³⁾. Effect of convection of UF₆ in liquid phase is treated as equivalent conduction heat transfer. Nusselt number is about 80.0, which was calculated according to the experimental formula of cavity fluid convection between surfaces of two concentric cylinders^{4),5)}.

Table 2 Material Properties of Carbon Steel

Density (kg/m ³)	7850
Specific Heat (kcal/kg °C)	0.11
Conductivity (kcal/m hr °C)	53.0

Table 3. Material Properties of Heat Protective Cover

	Temperature (°C)			
	162.	311.	475.	649.
Density (kg/m ³)	566.			
Specific Heat (kcal/kg °C)	0.12	0.22	0.23	0.24
Conductivity (kcal/m hr °C)	0.10	0.14	0.17	0.21

Table 4 Material Properties of UF₆

(1) Density and Specific Heat (s-in solid state, l-in liquid state)

	Temperature (°C)				
	10.	50.	64.(s)	64.(l)	150.
Density (kg/m ³)	5136.	4933.	4802.	3630.	3031.
Specific Heat (kcal/kg °C)	0.110	0.120	0.124	0.133	0.136

(2) Thermal Conductivity (s-in solid state, l-in liquid state)

	Temperature (°C)				
	20.	55.	64.(s)	64.(l)	
Conductivity (kcal/m hr °C)	0.353	2.579	3.439	315.0	

(3) Latent Heat

13.03 kcal/kg at 64.052 °C

3.6 VERIFICATION OF THE VALIDITY OF THE MODEL

For the purpose of verification of the validity of the analysis model mentioned in 3.3, some simple cases were conducted.

Figure 10 shows the object for the verification analysis. It is a steel cylinder whose length is 4.03 m, diameter (inside) 1.20 m and thickness of the shell 15.0 mm. Material inside the cylinder has similar thermal properties to those of natural UF₆, which is shown in Table 5. It occupies 60 % of the inside volume of the cylinder. As the thermal conductivity is set at infinity, temperature distribution of the material stays uniform.

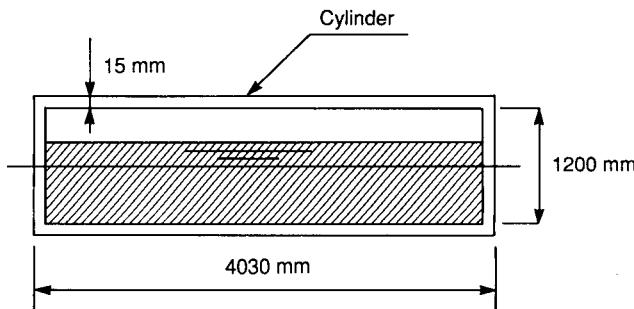


Figure 10 Cylinder Model for Verification Analysis

Table 5 Material Properties : Density is the value in solid phase. In liquid phase, it changes according to the temperature as is displayed in Figure 5.

Density (kg/m ³)	4800.
Specific Heat (kcal/kg °C)	0.120
Latent Heat (kcal/kg)	12.0 at 60.0 °C

In this verification analysis, three kinds of models were used. Two of them are axi-symmetric models which are equivalent to model-A and model-B mentioned in 3.3. The third one is a three-dimensional model shown in Figure 11.

Boundary conditions are similar to those mentioned in 3.4, under the fire accident condition of 800 °C for the duration of 30 minutes. In this verification analysis, both ends of the cylinder are insulated for the length of 715.0 mm from the edge shown in Figure 12 and effect of thermal gap between the material and the cylinder is not taken into account. For the three-dimensional model expansion of the material in liquid phase is considered.

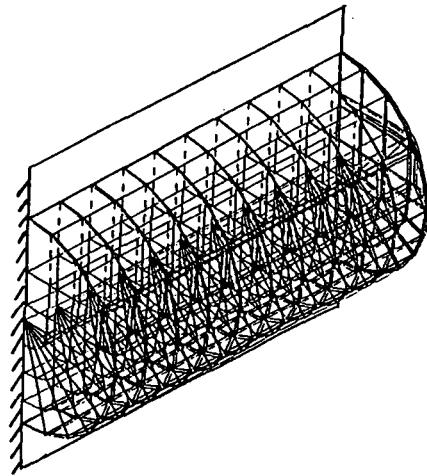


Figure 11 Three-Dimensional Model for Verification Analysis

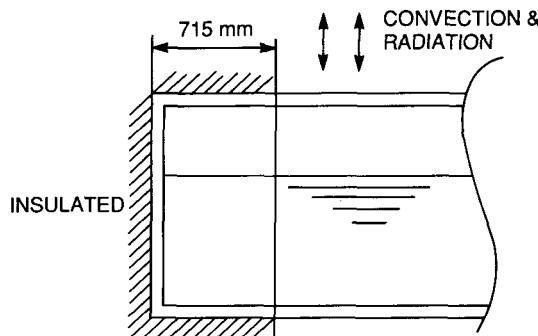


Figure 12 Boundary Conditions of Verification Analysis

Figure 13 and Figure 14 show the results of this verification analysis. The results of axi-symmetric models (model-A and model-B) and three-dimensional model show good agreement. And axi-symmetric models give conservative solutions.

Table 6 shows required CPU time measured on VAX 11/780 for the analyses. The method with axi-symmetric model displays excellent efficiency. The efficiency is about 45 times from the point of view of CPU time.

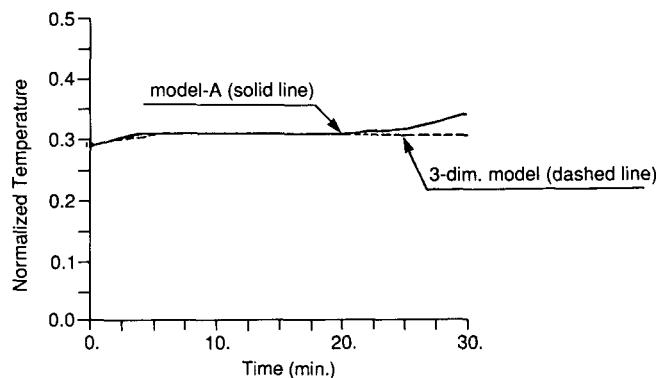


Figure 13 Results of Verification Analysis

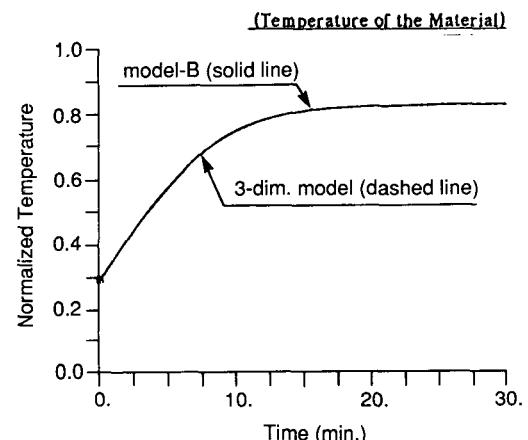


Figure 14 Results of Verification Analysis (Max. Temperature of Cylinder)

Table 6. CPU time of each model measured on VAX 11/780

model-A	380 sec.
model-B	100 sec.
Three-Dimensional Model	17500 sec.

3.7 RESULTS OF NUMERICAL ANALYSES

Table 7 and Figure 15-18 show the results of the numerical analyses of 48Y-cylinder filled with natural UF6.

Maximum volume of UF6 is 96% (CASE-1) and 86% (CASE-2) of cylinder capacity, so the hydraulic breaking does not take place even if without the heat protective covers.

Maximum vapor pressure is lower than critical internal pressure of cylinder in each case, so the breaking of cylinder by vapor pressure does not occur.

Maximum temperature of the valve is 144 °C in CASE-2 and the soft solder does not melt. Therefore the integrity of containment can be kept.

Table 7 Results of Numerical Analyses

	CASE-1	CASE-2
Maximum Volume of UF6	96% (45 min.)	86% (about 20 hr.)
Maximum Internal Pressure	24 kgf/cm ² (30 min.)	11 kgf/cm ² (30 min.)
Maximum Temperature of UF6	182 °C (30 min.)	146 °C (30 min.)
Maximum Temperature of Valve	144 °C (37 min.)	
Maximum Temperature of Cyl.	598 °C (30 min.)	595 °C (30 min.)
Critical Internal Pressure of Cyl.	28 kgf/cm ² (30 min.)	28 kgf/cm ² (30 min.)

*) Values in () indicate the duration after analysis has started.

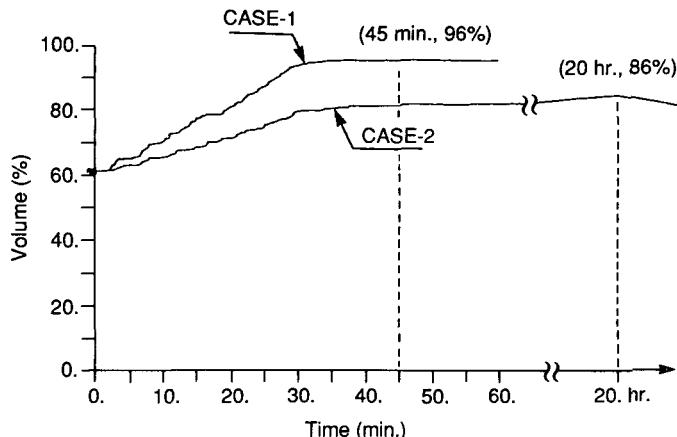


Figure 15 Results of Numerical Analysis (Volume of UF6)

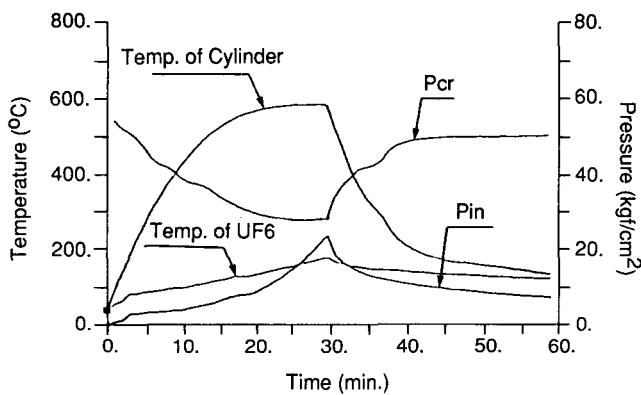


Figure 16 Results of Numerical Analysis (CASE-1)

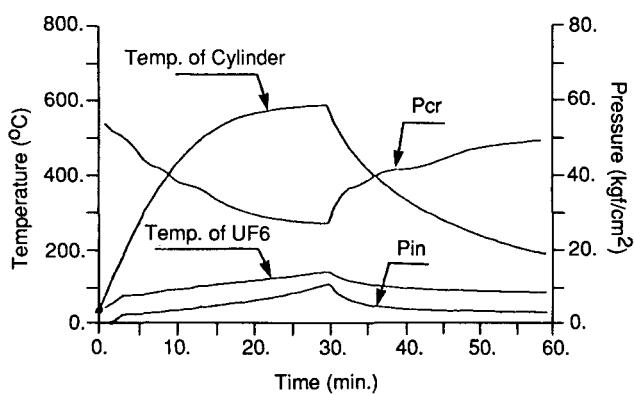


Figure 17 Results of Numerical Analysis (CASE-2)

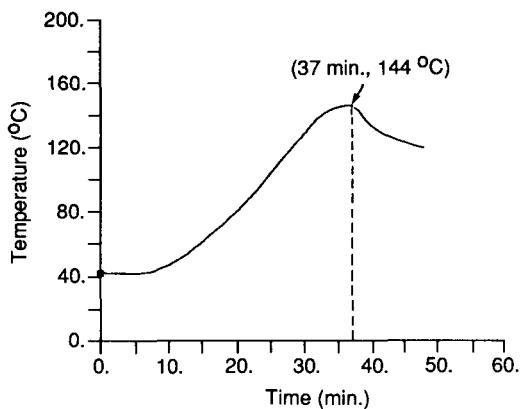


Figure 18 Results of Numerical Analysis (Temp. of Valve)(CASE-2)

4. REMARKS AND CONCLUSIONS

In this study, numerical analyses of 48Y-cylinder filled with natural UF6 were conducted under the condition of fire accident.

In this analysis, two kinds of simple axi-symmetric models were developed and safety evaluation was performed by using combination of results of the two models. The validity and efficiency of this method was verified by comparing the results with those of three-dimensional analysis.

Conclusions obtained from this study are as follows:

- (1) When 48Y-cylinder is filled with 12.5 ton of UF6, the hydraulic breaking does not take place under the fire accident condition of 800 °C for the duration of 30 minutes even if without heat protective covers.
- (2) Breaking of cylinder by vapor pressure doesn't occur at same condition as (1).
- (3) If heat protective covers are mounted, the soft solder of the valve doesn't melt.

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THERMAL BEHAVIOUR OF THE TYPE 30B CYLINDER EQUIPPED WITH THE 21PF.1 OVERPACK AND STUDY OF PROTECTIVE COVERS FOR THE 48Y CYLINDER VALVE

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ABSTRACT

This paper describes the tests which have been performed in France:

- first to verify the behaviour of 30B cylinders with their 21PF.1 protective overpack,
- secondly to develop a better mechanical protection of the valve.

1 - The thermal tests have been performed on real packagings filled with UF_6 , after the mechanical AIEA tests (free drop 9 m and 1 m on a pitch).

A methodical approach has been used with large thermal instrumentation:

- tests with inert material,
- transposition by calculation of the results to the case of UF_6 filling,
- confirmation tests with UF_6 .

The results were very satisfactory, the maximal temperatures which were measured on the external overpack of the 30B cylinder being much lower than the filling temperature.

2- In the Mont Louis wreckage, some of the valves of 48Y cylinders were damaged, as a consequence of the displacement of the protecting cover, and two of them were so damaged as to enable water to enter the cylinder.

A new valve protector was designed and was tested under impact situations.

The test programme was performed with the two designs: the standard one and the new protecting covers.

The tests have demonstrated that the new design which has the same weight as the original one and which may be used without any modifications of the cylinder, has a better behaviour, increasing the safety during transport.

INTRODUCTION

In the last few years, tests have been carried in France to check not only the mechanical and thermal behaviour of the 30B cylinders equipped with the 21PF.1 overpack, but also to improve the protection of the valve of the 48Y cylinder against the different loadings to which it can be subjected.

1. THERMAL BEHAVIOUR OF THE 30B CYLINDER EQUIPPED WITH THE 21PF.1 OVERPACK

1.1 General

The purpose of this study was to check the behaviour of the assembly consisting of the 30B cylinder, the 21PF.1 overpack and the sample bottle, particularly concerning the resistance of the cylinder to breaking when the package is submitted to drop and fire accident conditions with an actual UF_6 content, test conditions which had never been previously achieved.

The tests were carried out by the Commissariat à l'Energie Atomique (CEA) on the test facility of the Centre d'Etudes Scientifiques et Techniques d'Aquitaine (CESTA) near Bordeaux.

1.2 Nature of tests

The same cylinder-overpack-sample bottle assembly was subjected to three successive trials, with cumulative effect, in the following order [1]:

- free 9 metre drop onto a concrete slab covered with a steel plate, the impact occurred at the generatrix corresponding to the joint of the two half overpacks, in order to obtain damage corresponding to the maximum risk with regard to breaking in the subsequent fire test;
- free 1 metre drop onto a metal bar rigidly welded to the steel plate of the target, the impact occurred in the vicinity of the central reinforcement and the wooden frame of the overpack, the most fragile part of the zone already exposed the 9 metre drop, without opening the package;

- **fire:** after expert examination and instrumentation, the assembly already subjected to the preceding tests was placed 1 metre above the surface of a fuel source of kerosene on a specially designed stand; the joint of the two half overpacks was placed vertically so that the zone exposed to attack in the two preceding tests was exposed to the flames, the duration of the fire being 30 minutes.

Different phases of the tests were filmed from different angles for subsequent examination and checking.

For the fire test, the assembly was equipped with thermocouples connected to a temperature data acquisition system enabling monitoring and recording of data during test and subsequent cooling phase.

1.3 Test programme and results [2]

The final purpose of the test programme was to carry out tests on a cylinder filled with UF_6 but, for safety reasons, as it was preferred not to take the risk of releasing a large amount of UF_6 into the environment, nor to have a reaction between the UF_6 and the kerosene during the fire test, a methodical approach by steps was provided for:

- tests with inert ballast simulating UF_6 ,
- transposition by calculation to a test with UF_6 of the results obtained with inert matter, then examination of all experimental aspects from the safety viewpoint,
- tests with UF_6 taking the appropriate precautions.

1.3.1 Tests with inert ballast

The ballast used to simulate UF_6 consisted of a mixture of steel balls of 3 mm diameter and paraffin wax. This mixture was chosen because its physical properties are close to those of UF_6 : density, specific heat and thermal conductivity. Furthermore, the paraffin wax, which was introduced in liquid form at 60°C, is solid at ambient temperature and naturally sticks to the wall after cooling, making it possible to obtain a representative centre of gravity for the assembly, before the impact test.

After the 9 metre drop, the joint of the overpack was slightly distorted, the wooden frames of the two half overpacks being damaged. Two attachment bolts broke.

The puncture test with the bar resulted in substantial deformation and tearing of the overpack at the point of the impact.

On opening the overpack for examination and instrumentation for the fire test, it was found that the cylinder had turned through approximately 90° within the overpack, the only damage sustained was a dent 350 mm long, 200 mm wide and 25 mm deep caused by the bar.

For the fire test, the damaged package was instrumented with 28 thermocouples. Exposure to fire was extended to 90 minutes to study the safety margins.

During the first 30 minute period, the hottest point on the outside of the 30B cylinder was 75°C, with an average of 58°C for all the points of measurement; the sample bottle was at 25°C. After

90 minutes, these temperatures were respectively 170°C, 101°C and 70°C.

After complete cooling, the cylinder was immersed in water at a depth of 0.9 metre for 8 hours without water entering.

After these tests, the cylinder was submitted at 80°C to a hydraulic pressure of 28 bars without significant residual deformation.

1.3.2 Transposition of thermal results to fire test with UF_6

Using the preceding results, a calculation was made which showed that the temperatures reached by the cylinder in the test with UF_6 would remain, after 30 minutes, well below the temperature maintained at the time of filling (100°C) and that in the worst case, the amount of UF_6 melted would be very small.

The risk of hydraulic bursting could therefore be excluded, but leakage by the valve or plug remained a possibility.

1.3.3 Tests with UF_6

A new cylinder-overpack-sample bottle assembly was used. The 30B cylinder was filled with 2,150 kg of depleted UF_6 (U235 content 0.2%). The sample bottle, which was placed in its wooden box, contained 439 g of UF_6 .

1.3.3.1 Drop and puncture tests

The drop and puncture tests were carried out, as in the tests with inert ballast in the existing facilities, with the following results:

- **after the 9 metre drop test**
 - the joint between the two half overpacks was damaged and a 4 cm gap formed at the point of the impact;
 - no bolts were broken but 3 nuts at the joint, on the side opposite the impact, were loosened;
 - the outer metal skin of the upper half overpack had a tear 5 cm long close to the central angle section.
- **after the puncture test**
 - the outer metal skin of the overpack was substantially distorted and torn at the point of the impact.
- **after opening the overpack**
 - no damage to the sample bottle or its packing was found;
 - the 30B cylinder had turned through about 50° within the overpack;
 - there was a dent in the cylinder, caused by the bar, 485 mm long, 230 mm wide and 9 mm deep.

1.3.3.2 Fire test

For the fire test, a special area was fitted out. The damaged zone of the overpack was located at the bottom, exposed to the flames. The cylinder was placed so that the valve was at the top.

To detect any leakage of UF_6 from the valve and plug and to direct any outflow away from the kerosene pan, the cylinder was specially equipped. Leaktight lines were connected to the valve and drain plug; air circulation was provided in these lines which were connected to detectors.

The package to be tested was instrumented with 32 thermocouples (see Figure 1):

- 3 close to the overpack to measure the ambient temperature,
- 8 on the outer part of the 21PF.1 overpack,
- 9 on the inner part of the 21PF.1 overpack,
- 9 inside the 30B cylinder,
- 1 on the drain plug line,
- 1 on the filler valve line,
- 1 on the sample bottle.

After 30 minutes of fire:

- no leakage of UF₆ was found;
- the maximum temperature of the cylinder (see Figures 2 and 3) was 73°C, with an average of 43°C for the 9 points of measurement (a maximum of 75°C occurring 6 minutes after extinguishing the fire);
- the temperature of the lines, in the vicinity of the valve and drain plug, as well as at the sample bottle (see Figure 4) were respectively 34°C, 72°C and 16°C. The latter reached a maximum of 82°C in the subsequent phase, the wood contained in the ends of the overpack continuing to burn.

After opening the overpack it was found that:

- the 30B cylinder has suffered no damage during the fire test;
- the overpack joint on the site facing the fire was almost completely charred, as well as the greater part of the wood contained in the ends of the half overpacks;
- the protective wooden box and the sample bottle were intact.

1.4 Conclusion

The results obtained are satisfactory; the damage caused by the drop and puncture tests was very limited and did not affect the mechanical strength of the cylinder in any way. Also, during the fire, the maximum temperatures recorded inside the 30B cylinder remained well below the normal filling temperature, which eliminates any risk of breakage and shows that the cylinder equipped with its overpack could withstand considerably harsher temperature and duration conditions.

To comply with the tests specified for fissile packages, it would in addition be necessary to make allowance for the valve in the event of falling on one end, as although with regard to breaking of the container and the subsequent fire this could be considered to be a secondary aspect, in the event of a failure it could lead to leakage and the ingress of water after immersion.

2. STUDY OF PROTECTIVE COVERS OF 48Y CYLINDER VALVES

2.1 General

Type 48Y cylinders are used for the transport of UF₆, the uranium being depleted, natural or slightly enriched with a U235 content of less than 1%.

This packaging has a valve used for filling and draining. The valve is protected by a cover which, in the standard model [3], is secured to the skirt of the container with two set screws.

After the wreck of the Mont Louis cargo ship in the North Sea with 48Y cylinders filled with UF₆ on board, the cover was found to give inadequate protection against impact. A number of valves were bent, resulting in loss of the leaktightness in one cylinder. This damage was in most cases the result of displacement of the covers, most of them being removed.

After this incident, a new cover was designed in France with a view to improving protection of the valve in the event of impact. It was derived from those used for a number of years by COGEMA, COMURHEX and EURODIF at Pierrelatte for handling cylinders filled with liquid UF₆ at the sites. These companies, which have long experience of the use of packagings for UF₆, found that these new covers were satisfactory, giving better protection of the valve. The improved model of the cover consists in a frontal plate, a reinforcement bearing on the rounded bottom of the cylinder and a curved plate secured to the skirt of the cylinder by means of 4 set screws located inside the skirt (see Figure 5).

2.2 Description of tests

The tests were carried out at the COGEMA test facility at Moronvilliers, with a ballasted quarter-scale mock-up. As normal loading of a cylinder is not symmetrical with respect to the axis, the mock-up was provided with compartments to reproduce this asymmetry. The programme included two series of equivalent tests on packages equipped with standard and improved covers, in order to be able to compare deformation of the valve. In particular, the following tests were carried out:

- 0.60 m drops onto rigid slabs to meet the type A requirements, to which the cylinder could be subjected providing international agreement were obtained, if allowance is made for both the chemical risks (corrosive and toxic) of UF₆ and the presence of non-homogeneous deposits in many cylinders;
- 1.80 m drops, covering all possible interpretations of the regulations and also making it possible to assess the safety margin with respect to the preceding tests.

2.2.1 Tests on standard mock-up cover

- 0.60 m drop onto a slab, the axis of the mock-up being perpendicular to the plane of the target,
- 1.80 m drop onto a slab, the axis of the mock-up being perpendicular to the plane of the target,
- 0.60 m drop onto a slab, the mock-up being at an angle with the vertical passing through its centre of gravity and the edge of the skirt,
- 1.80 m drop onto a slab, the mock-up being at an angle with the vertical passing through its centre of gravity and the edge of the skirt,
- 0.10 m drop onto a bar with frontal loading of the cover,

- 0.30 m drop onto a bar with frontal loading of the cover,
- 0.10 m drop onto a bar with lateral loading of the cover.

2.2.2 Tests on improved cover

- 0.60 m drop onto a slab with the axis of the mock-up perpendicular to the plane of the target,
- 0.60 m drop onto a slab with the mock-up at an angle, the vertical passing through its centre of gravity and the edge of the skirt,
- 1.80 m drop onto a slab with the mock-up at an angle, the vertical passing through its centre of gravity and the edge of the skirt,
- 0.10 m drop onto a bar with frontal loading of the cover,
- 0.30 m drop onto a bar with frontal loading of the cover,
- 0.10 m drop onto a bar with lateral loading of the cover.

For frontal loading of the cover, the cylinder was slightly inclined with respect to the horizontal plane to allow the passage of the bar. The point of impact was located on the lower part of the cover (see Figure 6).

For lateral loading of the cover, the cylinder was at 90° with respect to the preceding test position. The cylinder was also slightly inclined to allow passage of the bar (see Figure 7).

2.3 Test results

For the different types of drop, the tests gave the following results:

- **drop onto slab**

For the two types of covers, the drops did not damage the valve.

- **drop onto slab at an angle**

Height of drop 0.60 m: with the standard cover, the valve was bent; with the improved cover, it remained intact.

Height of drop 1.80 m: with the standard cover, the valve was sheared off; with the improved cover, it remained intact.

- **drop onto bar**

Height of drop 0.10 m: the improved cover protected the valve in all cases, the valve remaining intact. However, with the standard cover, it was sheared off in the case of lateral loading.

Height of drop 0.30 m: in the case of frontal loading, the valve was damaged with both types of cover. It was bent with the standard cover and sheared off with the new model cover.

2.4 Conclusion

The tests confirmed what had already been concluded when assessing the safety of handling 48Y cylinders in the plants and when loading at incidents (the wreck of the Mont Louis cargo ship in particular), i.e. that the standard cover inadequately protects the valve against impact, particularly against lateral loads which can occur during handling.

In the event of a drop at an angle and impact on the valve, protection of the valve is considerably improved with the cover designed studied by COGEMA. Furthermore, as its weight remains within reasonable limits, being only slightly higher than that of the standard cover, it can be handled without special tackle and its use would necessitate no special modification of the 30B cylinders.

REFERENCES

- [1] IAEA Safety Series No. 6
- [2] Report on behaviour studies performed by CESTA (already forwarded to the competent American authorities)
- [3] ANSI Standard 14.1 (1982)

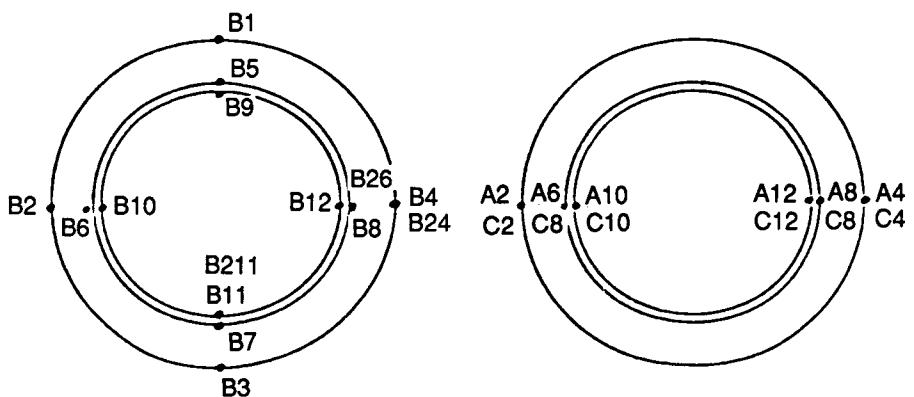
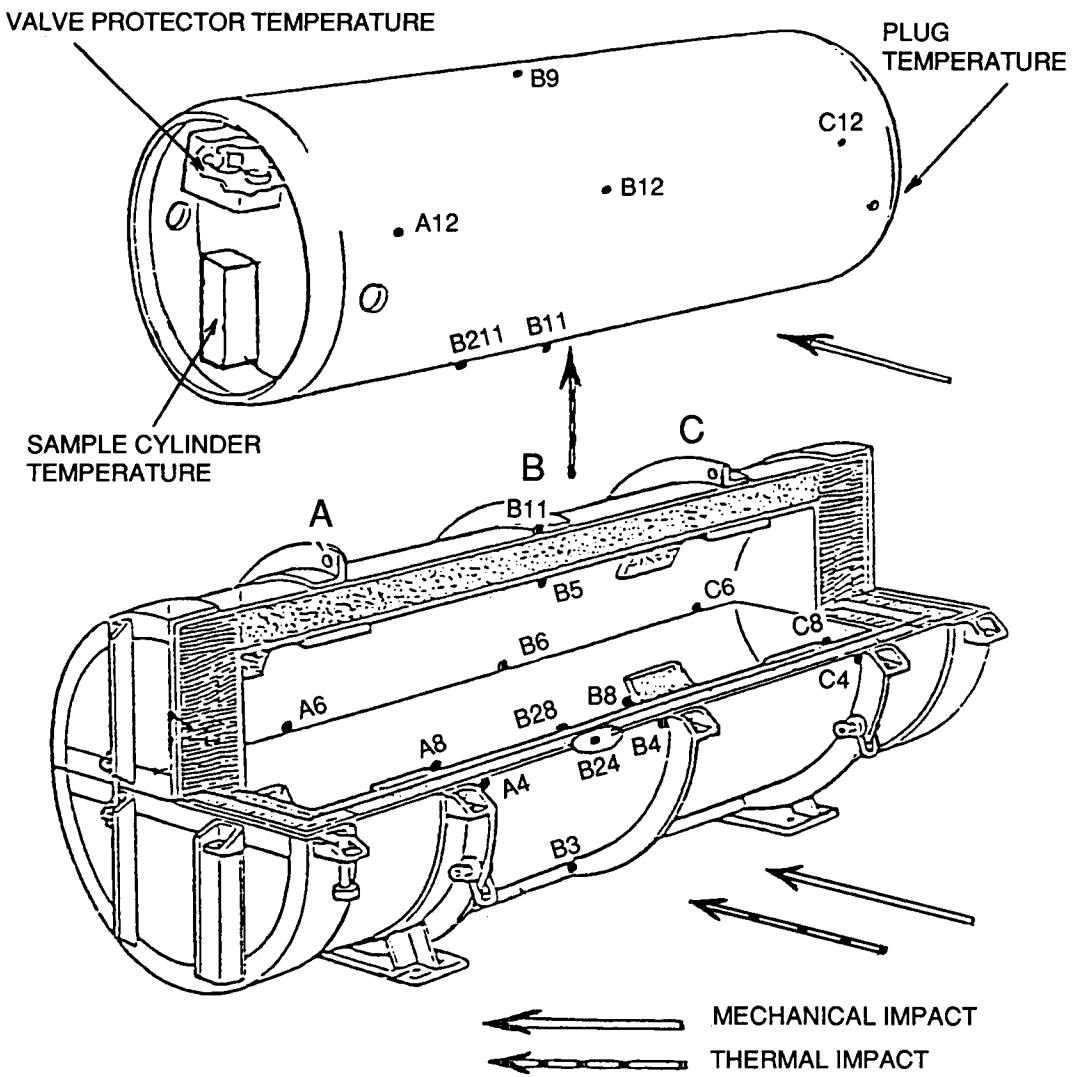


Figure 1

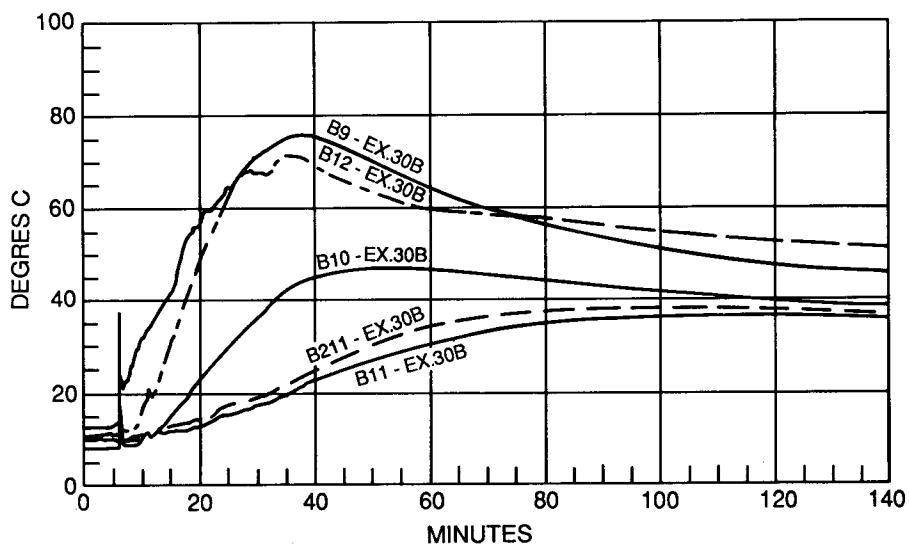


Figure 2

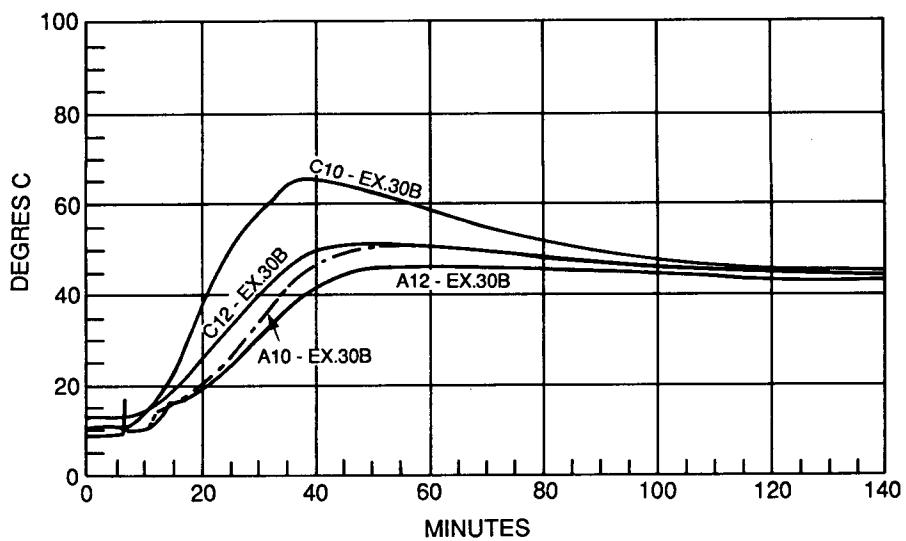


Figure 3

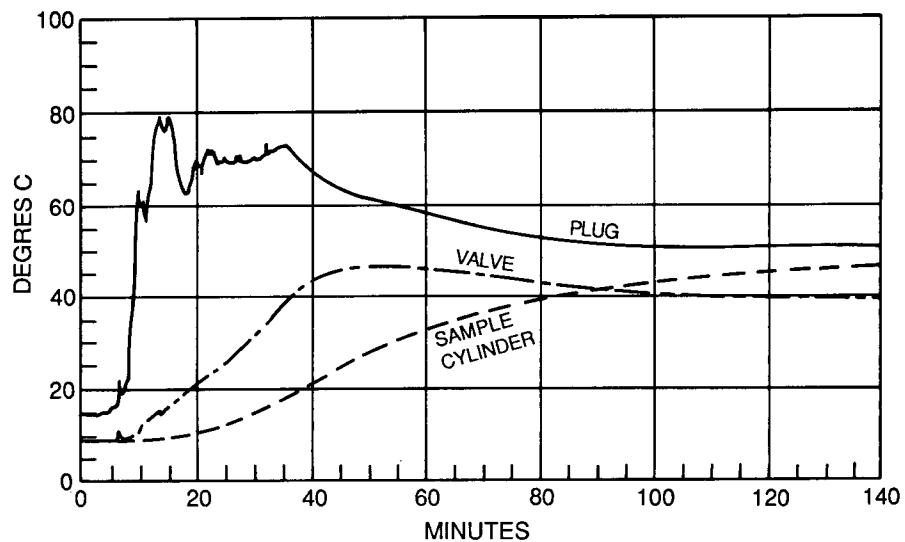


Figure 4

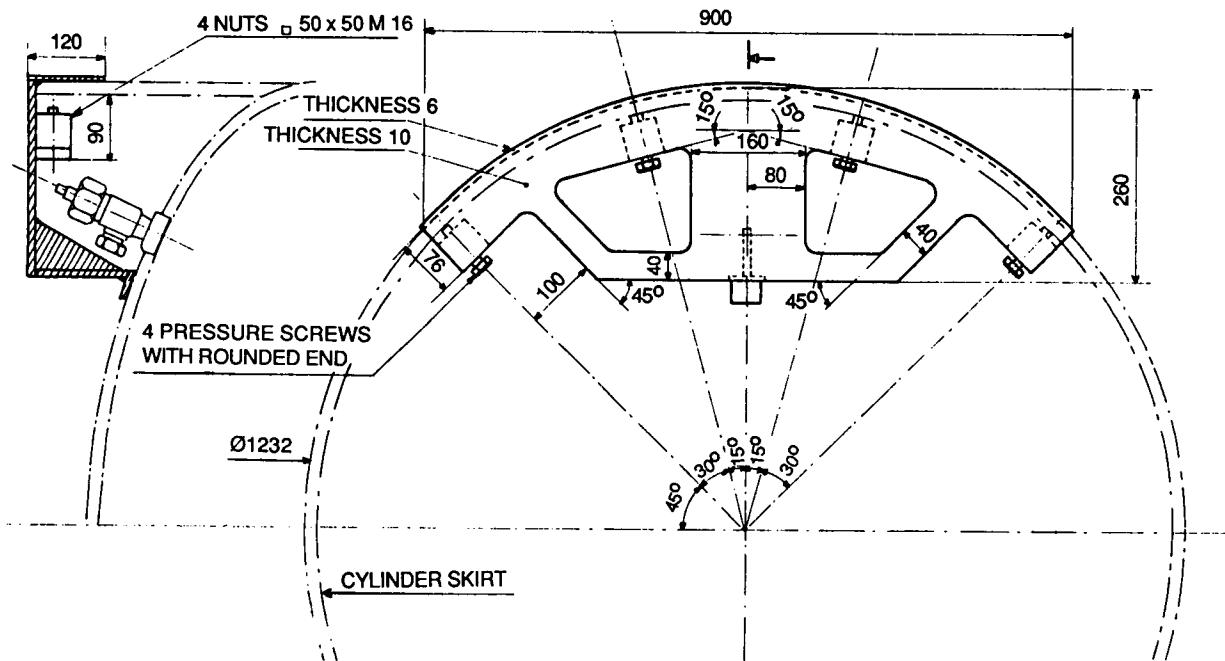


Figure 5

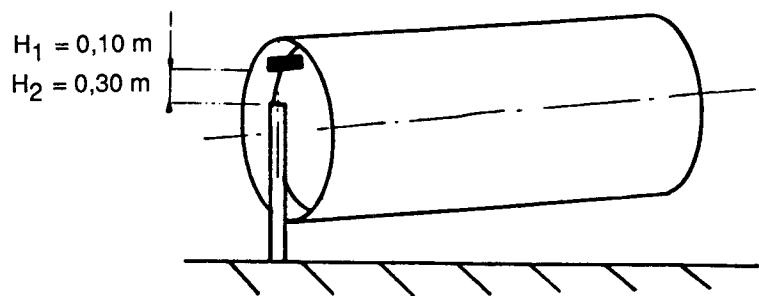


Figure 6

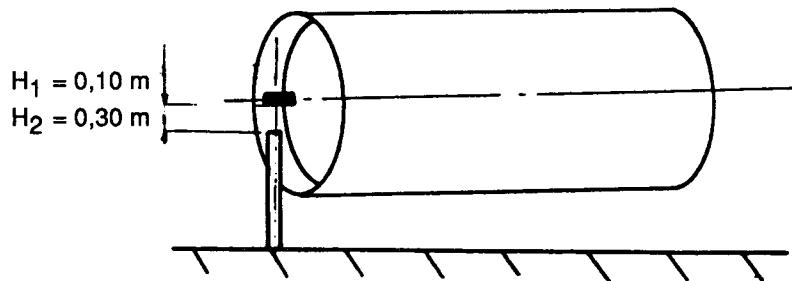


Figure 7

NEED FOR IMPROVED UF₆ HANDLING AND TRANSPORTATION PRACTICES

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ABSTRACT

Rigid design requirements for enriched uranium hexafluoride (UF₆) cylinder protective structural packages (PSPs or simply "overpacks" have contributed to the nuclear industry achieving greater than 30 years of operating experience with no transportation-related accidents resulting in the release of UF₆. However due to aging and continued over-the-road use, it is questionable whether all overpacks still meet the specifications to which they were manufactured. Overpacks are certified by their owners to be in conformance with U.S. Department of Energy (USDOE) ORO-651 and ANSI N 14.1 specifications; however, inspections often reveal this is not the case. Overpack maintenance seems to be done infrequently or not at all. UF₆ shipments from Portsmouth are not made until the existing regulations governing overpacks, tie-downs and the condition of trailers are satisfied. In numerous cases this has resulted in changing the initial shipment schedule.

Inasmuch as industry standards must be met, and the overpacks are not always given necessary maintenance, the need exists for more frequent inspections and routine maintenance.

INTRODUCTION

No fatalities or injuries have ever occurred due to accidents involving the transportation of UF₆. However, we must not become complacent or rest on this record. Safely transporting uranium hexafluoride for over 30 years has caused many to feel confident because of the many levels of safety intended to protect the public and the environment. The intent of this paper is to emphasize several safety related concerns that we at the Portsmouth Gaseous Diffusion Plant have as a result of our experience in shipping enriched UF₆.

DISCUSSION

The most important level of UF₆ cylinder protection is the packaging itself. All shipments of enriched UF₆ are made with the product cylinders encased in protective structural packages (PSPs) commonly referred to as "overpacks" which are the property of the licensed UF₆ user. The purpose of this overpack is to ensure that the cylinder will

withstand not only normal wear and tear but also the wide range of transportation insults that it might receive and still maintain its integrity.

Each overpack design is required to pass a series of tests before being accepted for routine use. The first tests are those associated with normal conditions of transport, whereas the second tests involve hypothetical accidents. Testing for normal conditions of transport requires exposure of the packaging to heat, cold, pressure, vibration, water spray, free drop within 2 hours of the water spray, corner drop, penetration, and compression. Testing for hypothetical accident conditions of transport involve a drop of 30 feet onto an unyielding surface; a drop of 40 inches onto an upright 5-inch by 8-inch bar; exposure to an environment of 1475°F for 30 minutes; and submersion in water for not less than 8 hours.

To ensure the continued reliability of the overpack, owners are required to provide certification accompanying each overpack which states that they have been inspected and that the overpack meets regulatory requirements. A signed statement from the owner is usually sent with each overpack; however, some overpacks are received that have been erroneously certified or have no certification and are not in an acceptable condition to transport cylinders of UF₆ product.

UF₆ shippers are required to inspect overpacks as specified in USDOE ORO-651 Revision 5, "Uranium Hexafluoride: Handling Procedures and Container Criteria". Section 6.4 states: "Protective overpacks shall be visually inspected by the shipper prior to each use. The following shall be cause for further investigation or removal from service until the defective condition is satisfactorily corrected: excessive warping, distortion or other damage of liner or shell which prevents a tight closure of the package; excessive clearances for inner container within the liner; fastener damage; reduction in thermal insulation thickness in any area; or any other damage or condition which would otherwise make the integrity of the protective overpack questionable as a fire-and shock-resistant housing. The vent holes should be inspected and resealed with an epoxy, if necessary, and the gaskets replaced or resealed, as required. The 30-inch protective overpack should be weighed periodically to determine if water has leaked into the overpack causing a

weight gain. Overpack tie-downs should be inspected to assure that they are not damaged and are adequate for their intended use."

The "American National Standard for Packaging of Uranium Hexafluoride for Transport," ANSI N14.1-1987, Section 7.4.1 also specifies a routine operational inspection. Besides including the foregoing from USDOE ORO-651, it states:

"The outer protective packaging shall also be inspected for evidence of the inleakage of water into the packaging. Any evidence of inleakage of water shall require an investigation of the packaging to determine the amount of water present in the packaging. The packaging may be required to have the weight recertified if found necessary by the investigation. The water shall be removed prior to repair of the outer protective packaging. Any nonconforming conditions found by the inspector shall be referred to personnel designated by the shipper to evaluate for the use, repair, or condemnation of protective packaging. The representative of the shipper shall contact the owner and user of the protective packaging for recommendations concerning any repair or modification of the packaging."

Inspection at Portsmouth indicates that many 30-inch cylinder overpacks no longer meet design criteria due to extended usage and normal wear and tear. Some owners have become lax in performing periodic preventative maintenance to the extent that an inordinate number of overpacks do not now meet design specifications. While it is the owner's responsibility to assure maintenance and repair of their own overpacks, the Portsmouth Gaseous Diffusion Plant occasionally performs some maintenance, which is billed to the owner, by repairing or replacing bad gaskets or stripped nuts and bolts, repairing minor welds, and repairing seal rings. We must reject overpacks when the repairs are extensive or exceed our repair capabilities. In that case, loading, and shipping schedules have been changed and the owner notified to send a replacement overpack. Examples of overpacks out of specification are shown in Figures 1, 2, and 3.

In addition to our concern with the questionable certification of overpacks discussed above, the receipt of cylinders and overpacks for international shipments has presented two other areas of concern -- tie-down methods and the condition of trailers. The first involves the various tie-down methods encountered for securing the overpacks to the shipping trailer beds. The design method for securing the overpack during shipment is to bolt its base to the trailer bed. Since most domestic shipping trailers are dedicated to UF₆ cylinder transportation, the overpacks remain bolted to the trailer beds and these overpacks are transported in the designed manner. However, numerous trailers for international shipment have been received with improperly and potentially dangerously secured overpacks. Examples of non-standard tie-down methods employed on incoming shipments are shown in Figures 4 through 7. In order that these shipments meet design specifications, holes will have to be drilled in the trailer bed and the overpacks properly secured for the return shipment.

The American National Standard Institute, is in the process of drafting Standard N14.2, "Tie-down

for Truck Transport of Radioactive Materials" and a subcommittee has been appointed to specifically address the methods of securing UF₆ 30-inch overpacks to trailers.

Tie-downs are recognized as an international problem and a conference addressed this topic on April 29, 1988 in Vienna. In addition, the International Atomic Energy Agency (IAEA) has issued for comment document ISO/DIS 7195 "Packaging of Uranium Hexafluoride for Transport." Section 6.2.3 is entitled "Tie-down Arrangements." The comment period is not yet closed, and it is unknown how soon or in what specifics the regulations will become effective. The IAEA has also issued for comment TECDOC-423, "Recommendations for Providing Protection During the Transport of Uranium Hexafluoride" which recognizes this problem area, but final review and implementation are yet to be determined.

The second major concern to have arisen with international shipments is the condition of the trailers themselves. Many trailers have not been loaded at Portsmouth due to mechanical problems, rotten flooring, bald tires, no brakes or brake lights, or broken springs. Examples of defects which have resulted in trailers being rejected for use are shown in Figures 8 and 9. A recent DOE directive states that questionable shipping practices are to be documented for DOE who will advise the DOT and appropriate state agencies.

CONCLUSION

Owners of cylinder overpacks must keep current their required periodic inspections and maintenance to assure that shipment schedules are not changed due to rejection of the overpacks and more importantly, to ensure that the safety of the cylinder is maintained during transport. The Department of Transportation Research and Special Programs Administration (RSPA) has issued for comment docket HM-166V, "Hazardous Materials, Uranium Hexafluoride" which addresses periodic overpack maintenance and which will codify ANSI N14.1 1987.

Currently there are no regulations for the method of securing overpacks, however, this need is being addressed by ANSI and other organizations. Our experience strongly suggests that when regulations and standards are received, they must be implemented expeditiously to guarantee maintaining our safety record. Even though these regulations will be a most positive development when finally issued, I further propose that a program be established to design special trailers and dedicate them to the transport of UF₆ nationally as well as internationally. Engineered tie-downs can be developed on a low center of gravity trailer whose maintenance can be monitored and whose owners would be held accountable for trailers meeting DOT standards.

Our industry has compiled an enviable transportation safety record due to the basic design integrity of the packaging. By developing dedicated trailers we will further demonstrate our continued commitment to safety and control of all facets of the transportation cycle. Together we must continue to fulfill our transportation and logistics responsibilities, and maintain our enviable safety record in the handling and transportation of UF₆.

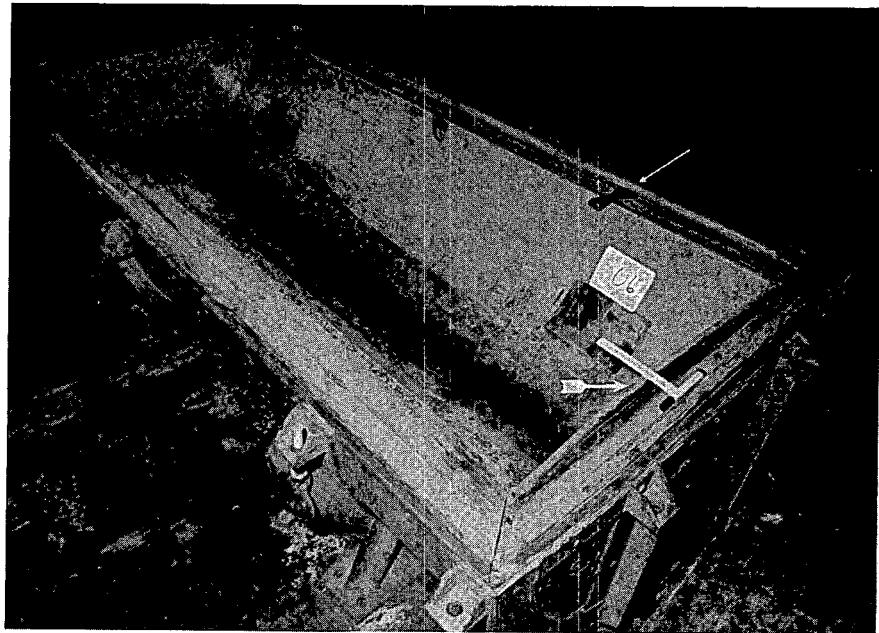


Fig. 1. Overpack Interior Showing Water Inleakage, Loose Strip Joint (Arrow at Knife Under Stop Strip), and Warped Steel Liner (arrow)



Fig. 2. Overpack Interior Showing Disintegration due to Water Saturation of the wooden Frame Member

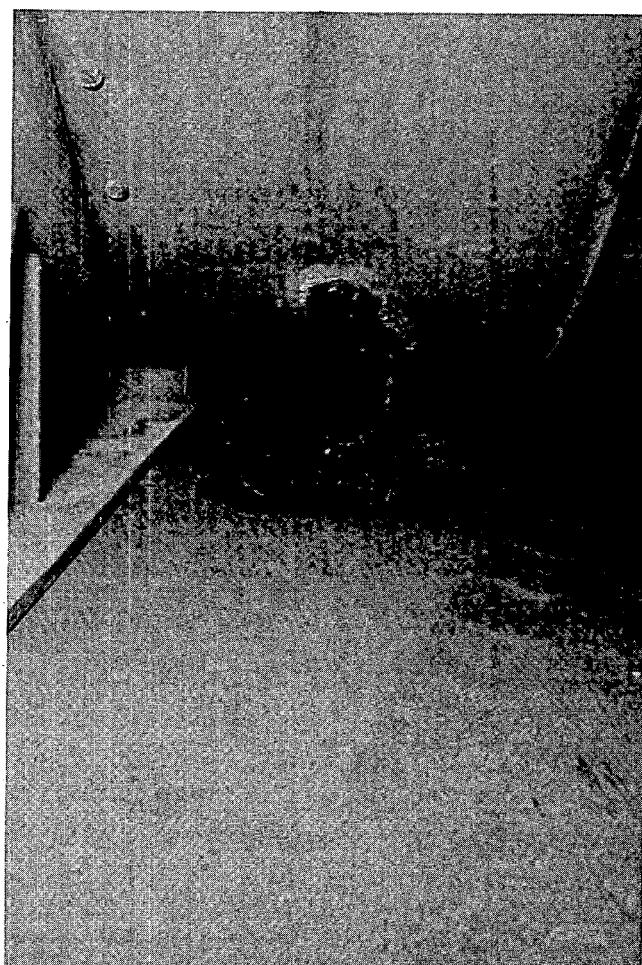


Fig. 3. Leakage of Water Through the Overpack Exterior Wall



Fig. 4. Overpack-Trailer Tie-Downs Employing Two $\frac{1}{2}$ " Steel Cables

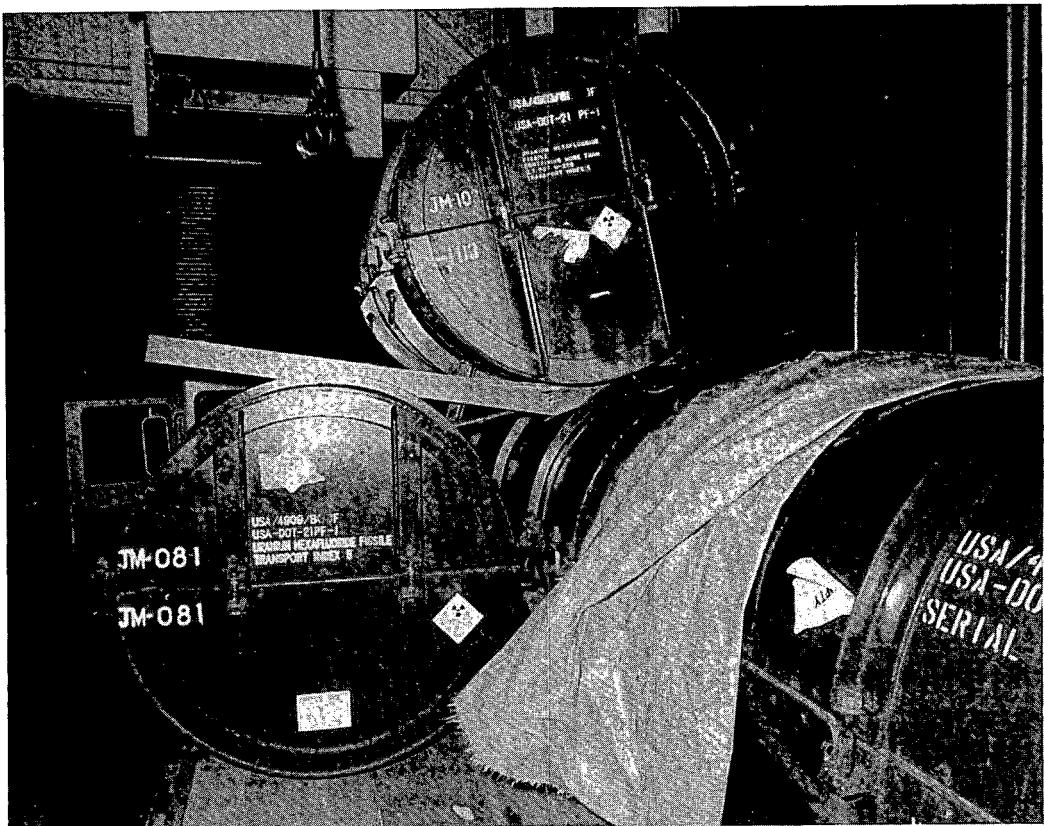


Fig. 5. Example of Overpack Loading Practice

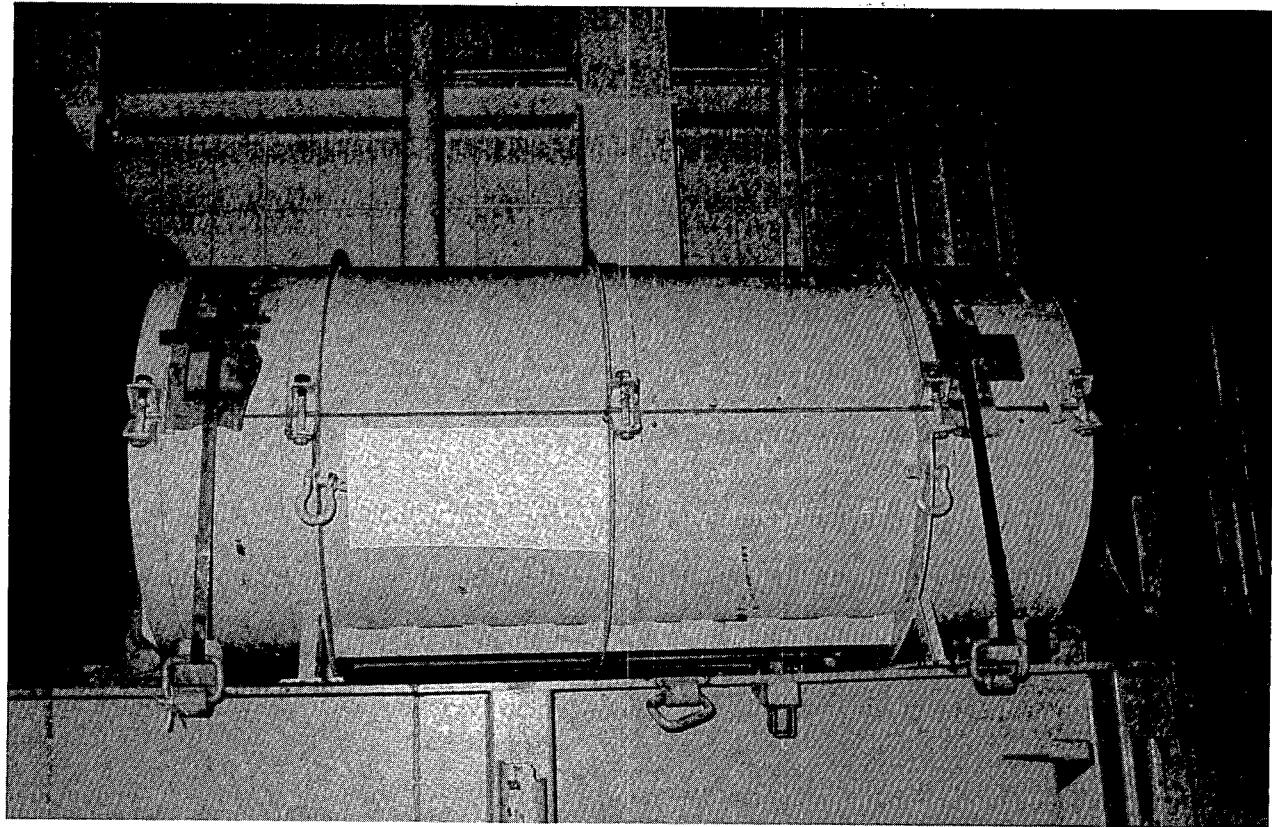


Fig. 6. Overpack-Trailer Tie-Down Employing Two Steel Straps per Two Overpacks

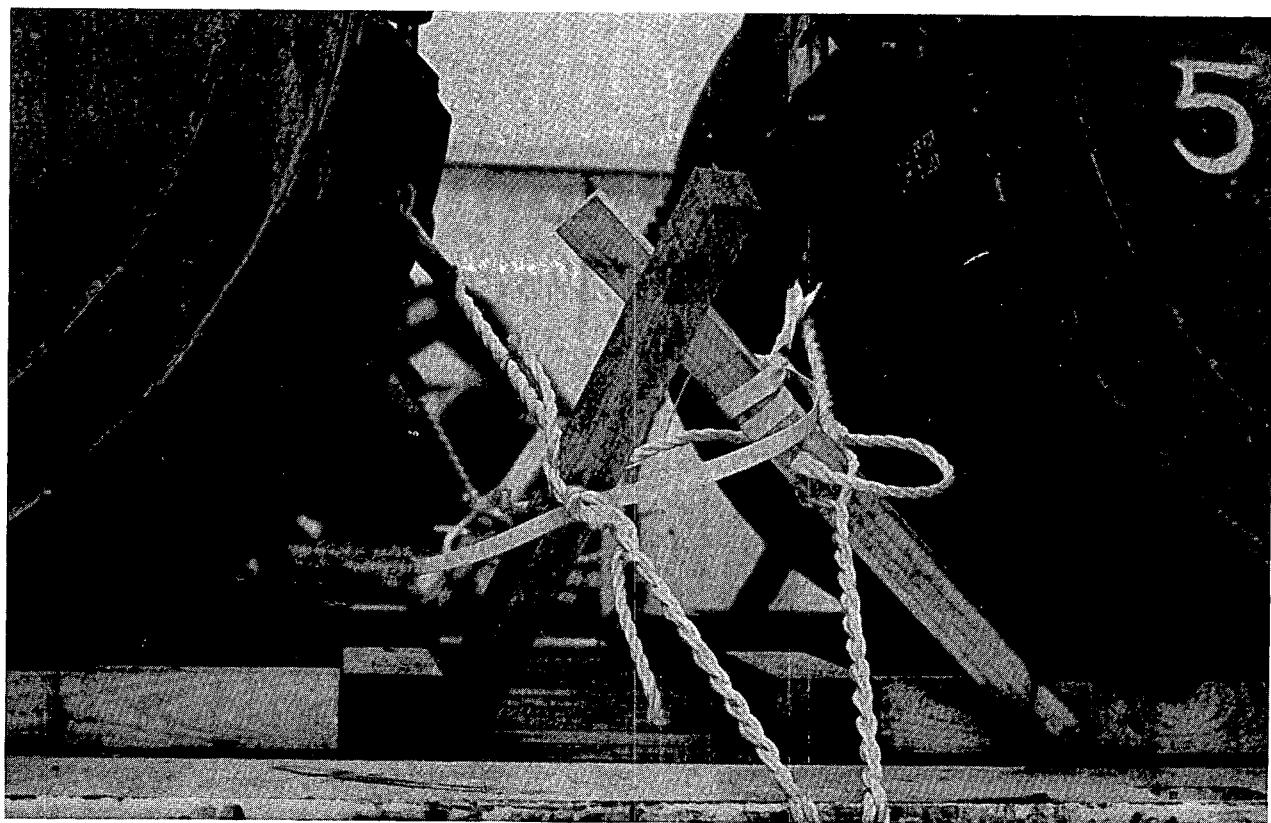


Fig. 7. "Boy Scout" Overpack Tie-Down Method

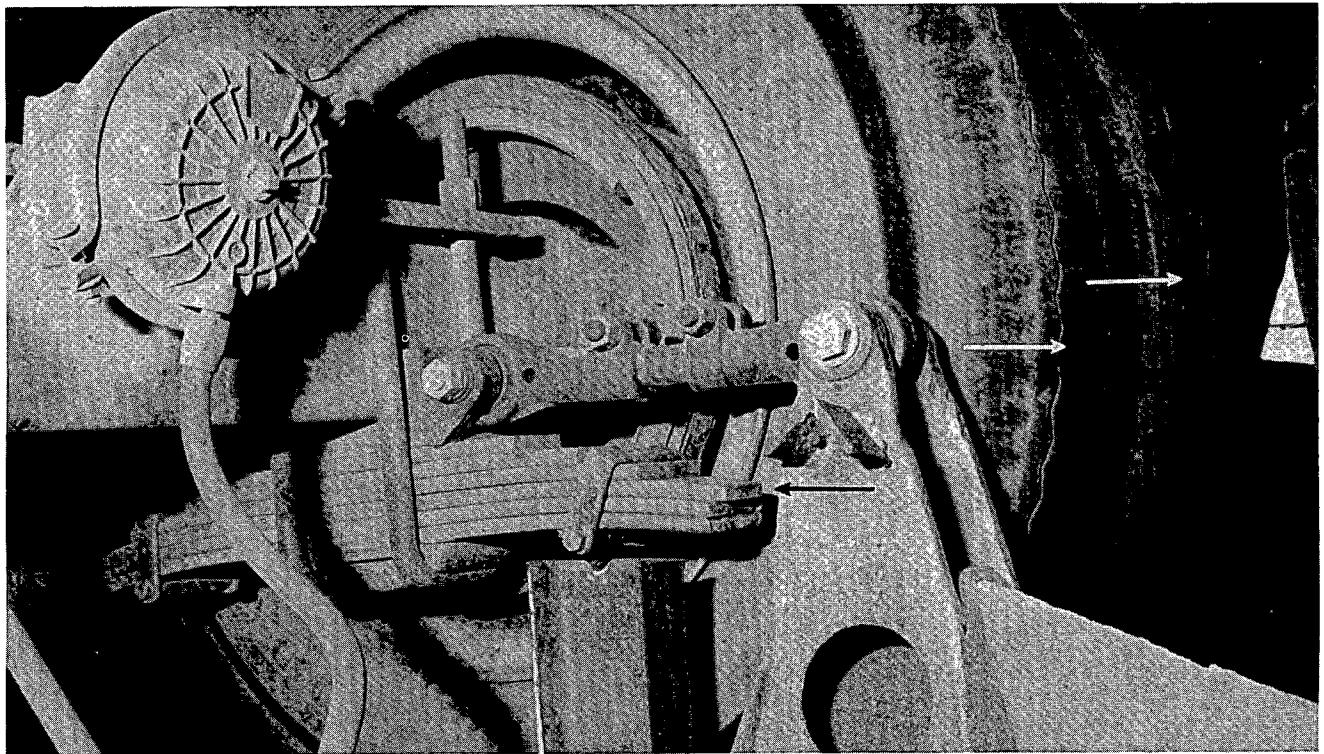


Fig. 8. Overpack Trailer Showing Broken Spring (arrow) and Bald Tires (arrows)

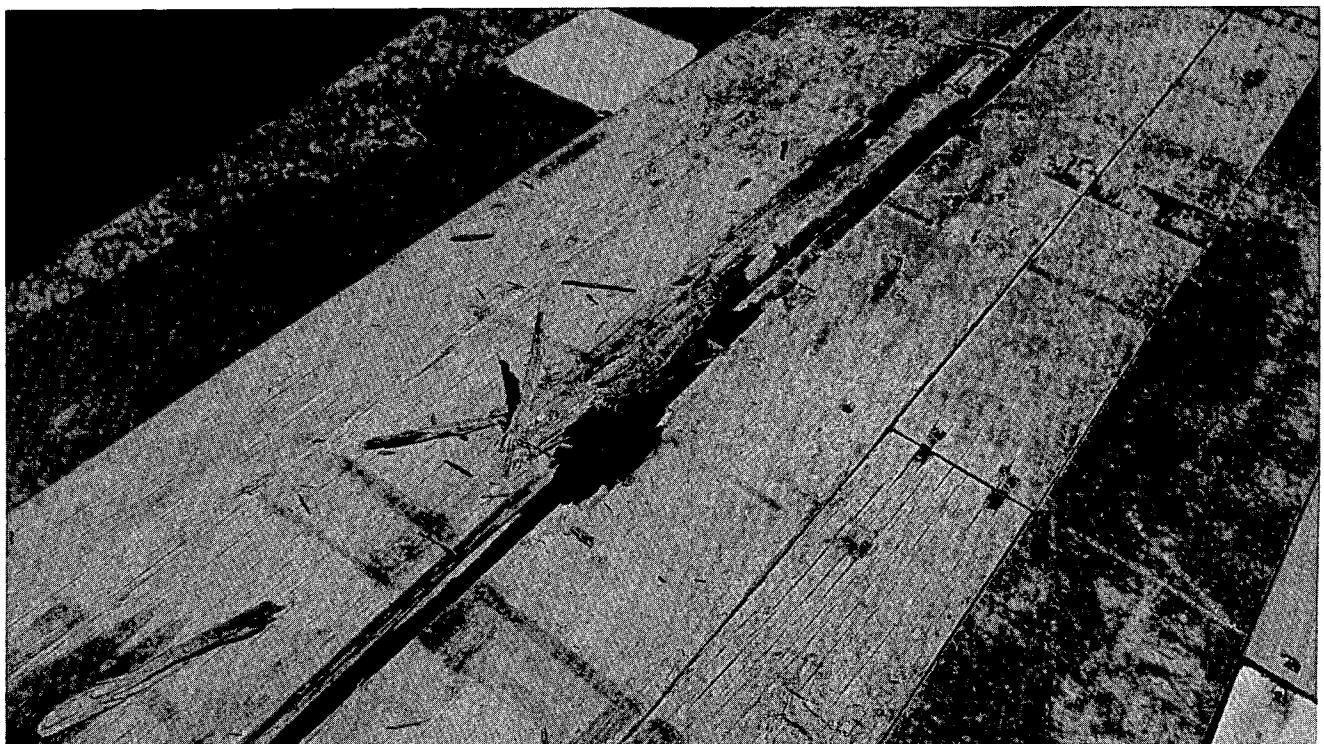


Fig. 9. Overpack Trailer Bed Showing Deteriorated Planking

Prepared by the Portsmouth Gaseous Diffusion Plant, Piketon, Ohio, 45661, operated by MARTIN MARIETTA ENERGY SYSTEMS, INCORPORATED, for the U.S. DEPARTMENT OF ENERGY, under Contract No. DE-AC05-76OR00001.

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COMPLIANCE ASSESSMENT OF AN URANIUM HEXAFLUORIDE PACKAGE 30B
WITH OVERPACK TO THE IAEA STANDARDS.

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ABSTRACT

At the Dipartimento di Costruzioni Meccaniche e Nucleari (DCMN) of the Pisa University a research program was carried out in order to assess the compliance to the updated IAEA standards of the UF6 30B container, complete with its sandwich phenolic foam filled external overpack.

The research program, performed in collaboration with ENEA and several interested Italian firms, included 9 mt free drop, perforation, thermal and leaktightness tests, on two complete packages with dummy load.

The heat transfer conditions, with the UF6 real contents, were simulated by means of numerical analyses with the TRUMP computer code and calculation procedures set up using the available experimental data.

The attained results seem to be useful from the point of view of the foreseen purposes.

1. INTRODUCTION

In Europe and in particular in Italy the UF6 transport is carried out mainly by means of containers and casks designed in the USA and locally built according to the relevant original specifications /1,2,3/.

In the recent past in Italy the most used UF6 cask for transport and/or interim storage purposes was the 30B container complete with a phenolic (foam filled overpack, necessary because of the U235 contents).

After the expiry of the previous license released by the Italian Authority on the basis of the USA technical documentation, taking in account of the quite large number (over 1000) of specimens then in use in the country, it was felt necessary to acquire information useful for a possible assessment of the cask model reliability with reference to the up to date IAEA standards /4/.

Therefore a research program was set up and carried out at the Dipartimento di Costruzioni Meccaniche e Nucleari (DCMN) of the University of Pisa in collaboration with ENEA and national firms interested in the cask use, manufacture and/or transport (i.e. ENEL, AGN, HOVAL, BORGHI), within the frame of the ENEA safety research programmes.

In fig. 1, a UF6 package schematic drawing is shown.

2. RESEARCH AND ANALYSIS PROGRAM

The foreseen program included the following tests, in the IAEA standard conditions, on two specimens of the package with casting granulate dummy load:

a) First specimen:

- Preliminary geometrical and leakage rate checks
- 9 mt lateral drop test (package axis horizontal)
- Perforation test (1 mt drop on a cylindrical bar with impact against the overpack lateral surface)
- furnace test

b) Second specimen:

- Preliminary geometrical and leakage rate checks
- 9 mt corner drop test (with package axis inclined by 27° on the vertical)
- 1 mt drop test against a cylindrical bar (impact against the overpack bottom)
- 9 mt lateral drop test
- Perforation test (impact against the overpack lateral surface)
- furnace test.

Before and after each test the 30B cask leakage rate by means of drop pressure tests (or alternatively Helium tests) were measured. Moreover the program included also the analysis of the behaviour of the package with the UF6 real contents in the IAEA thermal test conditions, by

means of calculations performed with the FD TRUMP code set up on the available test data.

3. TEST RESULTS

3.1 - Mechanical test results

In the fig. 2 the first package before the 9 mt lateral drop test is shown.

In this test the package was lifted with his axis horizontal in order to localize the impact on the overpack two half - shell closure.

The overpack deformations are shown in the fig. 3.

An identical test was performed on the other specimen with similar results.

The fig. 4 shows the overpack permanent deformations measured on the two packages after the lateral drop test.

During the drop test the 30B cask accelerations were recorded.

In the fig. 5, the 30B accelerations (as recorded), the displacement and velocity, obtained by double integration, are plotted versus the time. The maximum registered acceleration and impact duration in the two lateral drop tests have been respectively 350 g and 19 msec.

On the second package also a 9 mt corner drop test was performed. The fig. 6 shows the package configuration before the lifting. The package damage and the permanent deformation values are indicated in figg. 7 and 8. The plastic deformations are concentrated in correspondence of the impacted edge on the overpack as well as the 30B cask.

The fig. 9 shows the acceleration diagrams registered during the test in direction normal and parallel to the package axis respectively.

The maximum acceleration and impact duration in this case were 132 g and 296 msec respectively.

Both the specimens were submitted to the IAEA perforation test (1 mt drop test against a cylindrical bar) concentrating the impacts on the overpack cylindrical surface in a zone far from wood ribs.

The figg. 10-12 show the first specimen before the test and the overpack and 30B damages. In the fig.13, the overpack and 30B permanent deformation on the overpack lateral surface are reported. The second specimen was submitted also to a perforation test on the overpack bottom in the area of the 30B valve.

The fig. 14 shows the overpack damages and the measured deformation values.

As result of the impact on the bar, the valve housing was "forged" against the valve body and the package leakage rate (see par. 3.3) increased quite largely.

3.2 - Evaluation of the package thermal behaviour

3.2.1 - Thermal tests

As it was foreseen in the general program, the thermal behaviour of the package in the reference accident conditions (i.e. furnace tests) was assessed mainly by means of calculations performed with the FD code TRUMP /7/.

The suitability of the code and calculation procedures were tested by means of two IAEA standard furnace tests on the same specimens (with the simulate contents) used in the drop and perforation tests.

The "furnace tests" were performed in a quite large gas furnace normally used for heat treatment of welded structure in a factory near Pisa.

For the tests the specimens were introduced in the furnace (fig.15) previously heated overnight at over 850° C.

After the 30' test the specimens were withdrawn from the furnace and allowed to cool down in air (actually to keep burning) unattended for three hours.

Even if it was not possible to obtain a complete temperature pattern of the packages during the tests, mainly because of partial transient damage of the experimental set up due to the industrial environment and the characteristics of the furnace, experimental data on the maximum value as well as information on the temperature trends in the heating and cooling time were achieved by means of Thempil melting indicators (in the 120°-430° C range) and Chromel-Alumel thermocouples connected to a digital graphical AUTODATA - 616 SCANNER ACUREX data acquisition set.

The main results may be summarized as follows:

- On the surface of the 30B container, maximum temperatures of about 220°C (conservative value) were reached under the lateral perforation caused by the mechanical tests and in the surrounding areas;
- On the two bases of the 30B container the maximum temperature in both the tests reached values over 120°C and below 220°C;
- On the surface and in the bulk of the dummy contents maximum temperatures of about 70°C and 35°C were reached respectively after 1 h of cooling time at the end of the test (210').

3.2.2 - Numerical analysis

The numerical simulation of the tests required several calculation runs by means of the TRUMP code and the FED processor /5/ including:

- 1) Preliminary approach with a triaxial mesh regarding about a 1/8 of the package .
- 2) Calculations, regarding a section of the 30B container around the perforation mark axis with several rather detailed meshes and material characteristic derived directly or calculated by means of correlations found in pertinent reference (i.e. /6/, /7/, /8/).

As it is possible to see in the fig. 16, a) the values and the trends of the available test and

calculated data for the 30B container seem to be in an rather acceptable agreement.

For the thermal analysis of the package with the UF6 contents, the calculations were performed in the same conditions and hypotheses used in the test simulation with other assumptions related to the initial temperature of the package (38°C) and a conservative behaviour of the UF6 (adiabatic vaporization/condensation in the 30B container; evolution of the phase changements through equilibrium situations, etc.).

In the fig.16 b the diagrams of the temperature in several points of the 30B container are shown.

In any case the pressure, corresponding to the UF6 maximum calculated temperature (80°C) under the penetration mark, results to be acceptable for the structural integrity of the 30B container.

3.3 - Leaktightness Test Results

The damages caused by the tests to the package leaktightness was assessed by means air pressure drop tests. For this purpose, the 30B container was pressurized up to 1 bar with air. The experimental set up used allows to obtain the leakage rate measuring the pressure drop and the cask and test environment temperatures at constant time increments. The registered data are elaborated by means a computer code which performs the statistic elaboration and check of the test data and calculates the leakage rate. The demonstrated method sensitivity is 10^{-4} mbar l/sec.

In tab. I the 30B leakage rates obtained by the several leaktightness tests performed after each mechanical and thermal test are reported. The first specimen initial leakage rate resulted to be equal to $1.14 \cdot 10^{-2}$ mbar l/s. This figure was not practically affected by the performed mechanical tests moreover the thermal test seems to decrease the leakage rate by a factor 2. This result can be explained by the reduction of the gaskets microscopic porosity due to the products of the foam and wood combustion.

The initial leakage rate of the second specimen was about $5 \cdot 10^{-2}$ mbar l/s. After the corner drop and penetration test in correspondence of the valve position the measured value was 19.8 mbar l/sec. Screwing in further the plug valve (with a torque equal to 7 Kgm) the leakage rate decreases to 3.038 mbar l/s. The leaktightness test performed on the second specimen after the replacement of a new valve showed again that the lateral 9 mt drop and perforation tests, as well as the furnace test don't affect the cask leaktightness.

The leakage rate registered after the 9 m corner drop and the performance tests (leaktightness test n.5 in tab. III) don't produce criticality problems. In fact in the IAEA immersion test (par. 633/4/) with the actual 30B inner pressure condition ($p = 0.2$ bar), the water in leaching in the cask was evaluated to be about

80 cm^3 on the basis of immersion tests performed with the damaged valve mounted on a smaller test container. On the contrary calculations performed by AGN indicated that criticality risk might arise with a water in take of about $1.4 \cdot 10^5 \text{ cm}^3$.

4. CONCLUSIONS

The results attained in the research program seem to lead to several conclusions that may be useful for a possible assessment of the reliability of the package in particular:

- 1) The most severe mechanical test appears to be the penetration one that may impair slightly the leaktightness the loading/unloading valve and produce the complete perforation of the overpack.
- 2) Even though the eventual dispersion of the contents, does not induce radioprotection problems, the possibility of chemical pollution hazards and minor water in-leaching events might be eliminated by quite simple modifications to the valve cover.
- 3) The temperatures attained in thermal test conditions on the 30B inner container around the perforation area appear not dangerous for the package structural integrity.

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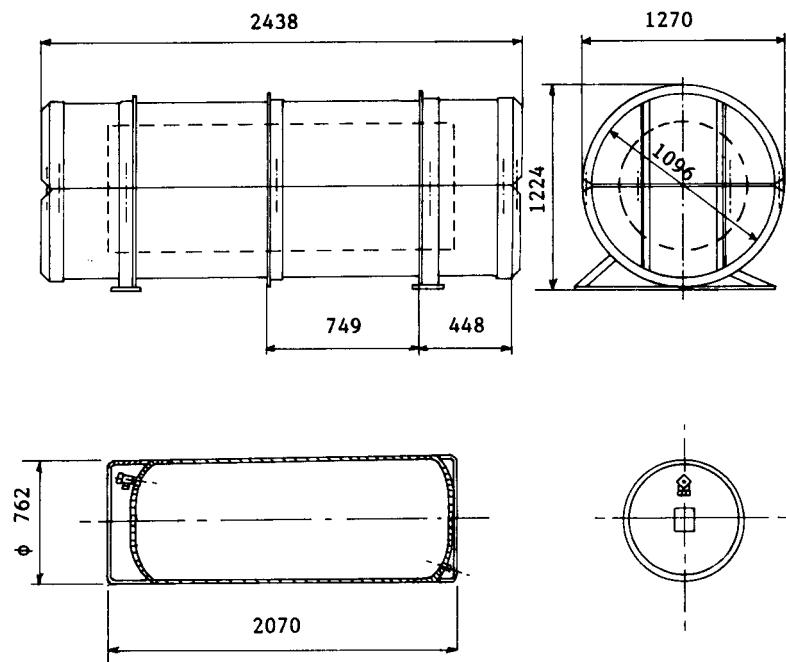


Figure 1. UF6 package schematic drawing

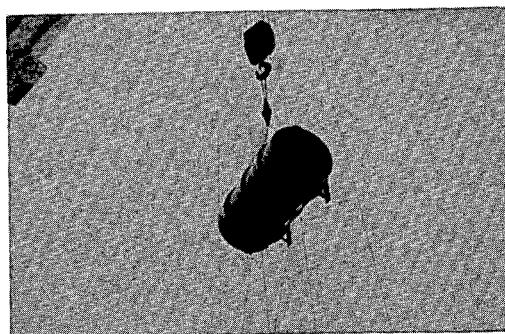


Figure 2. UF6 Package 4561/BTN 2103 before the lateral 9 mt drop test

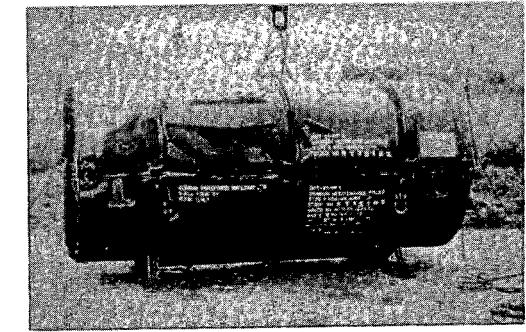
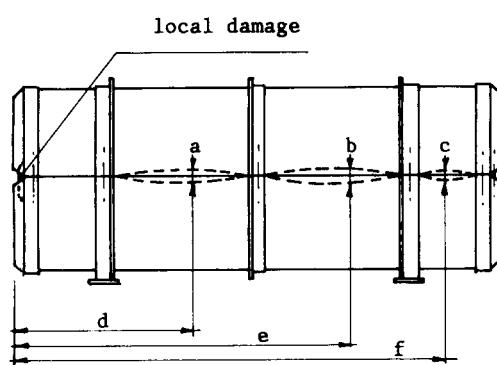


Figure 3. Damage of the 4561/BNT 2103 package due to the 9 mt lateral drop test



package specimen	a (m)	b	c	d	e	f
4561	45	50	25	840	1450	2098
4563	49	52	29	900	1655	2110

Figure 4. Overpack damages caused by the 9 mt lateral drop tests

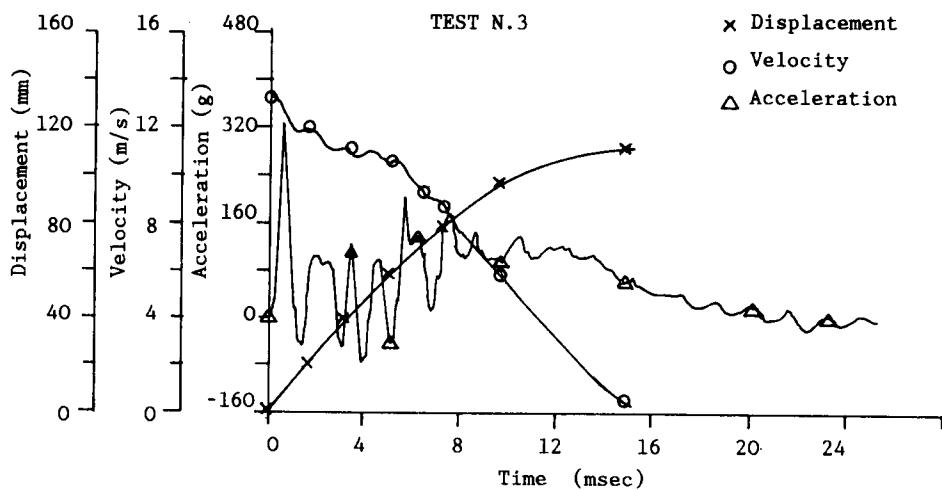


Figure 5. Acceleration registered during the 9 mt lateral drop test

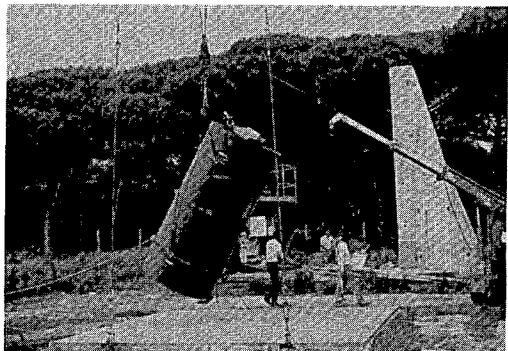


Figure 6. UF6 Package 4563/BTN 2094 before the 9 mt corner drop test

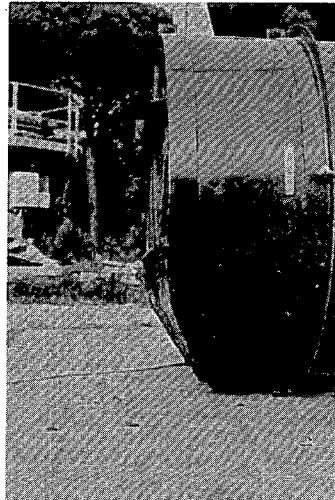


Figure 7. Overpack damage caused by the 9 mt corner drop test (package n. 4563/BTN 2094)

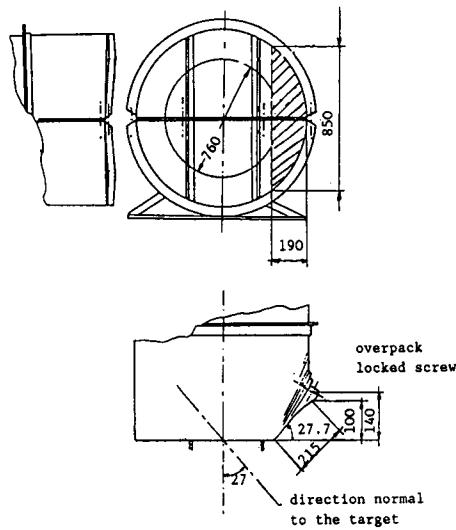


Figure 8. Overpack damage caused by the 9 mt corner drop test (package n. 4563/BTN 2094)

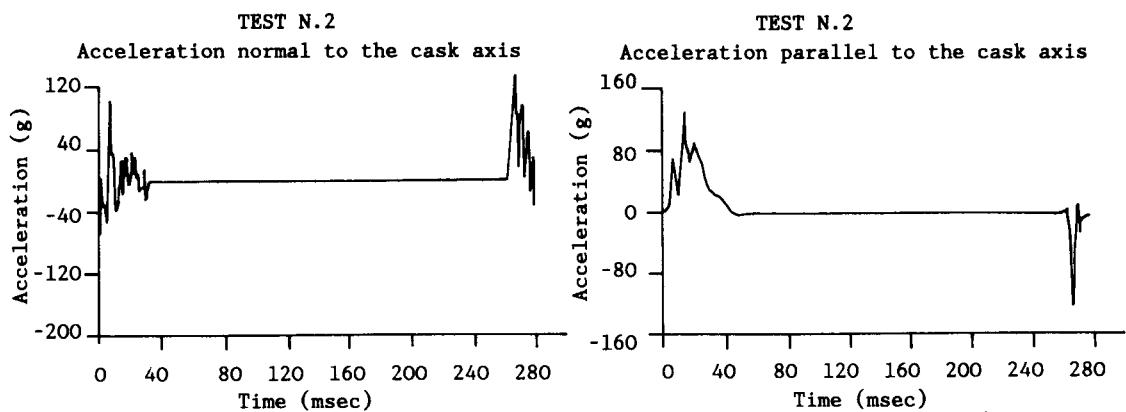


Figure 9. Accelerograms registered during the 9 mt corner drop test (package n. 4563/BTN 2094)

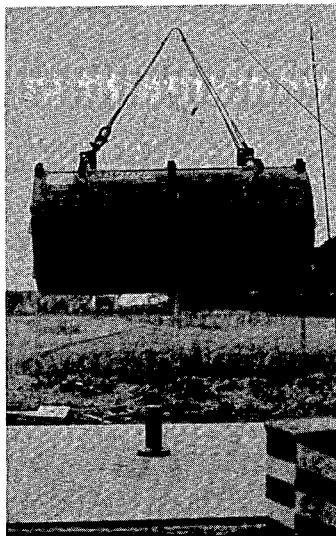


Figure 10. Package 4561/BTN 2103 before the perforation test against the overpack lateral surface

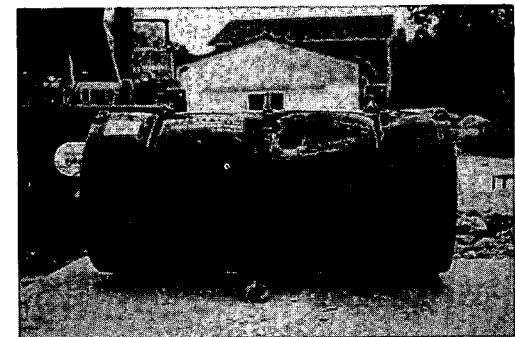


Figure 11. Overpack damage caused by the lateral perforation test (package n. 4561/BTN 2103)

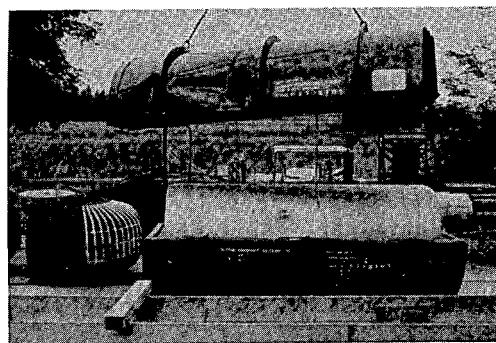
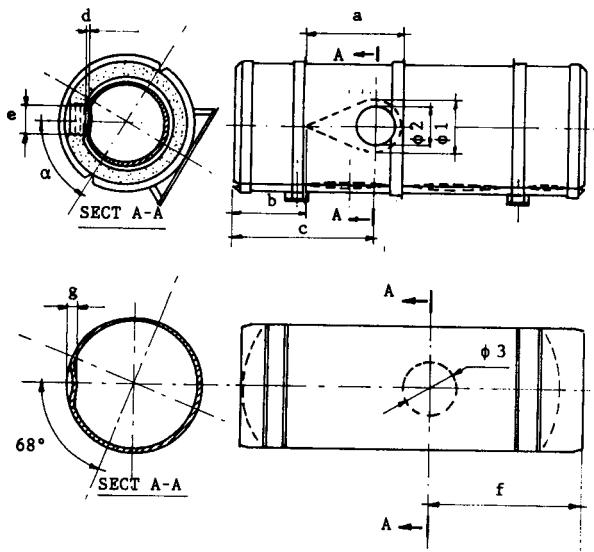


Figure 12. 30B cask damage caused by the lateral perforation test (package n. 4561/BTN 2103)



Package specimen	α	ϕ_1	ϕ_2	ϕ_3	a	b	c	d	e	f	g
4561	22°	350	165	350	752	490	1010	12	155	905	35
4563	316°	180		320		532	962			875	32

Figure 13. Overpack deformation values caused by the lateral perforation tests

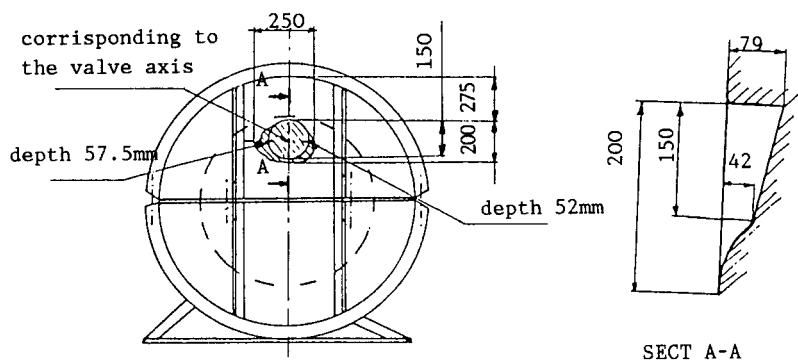
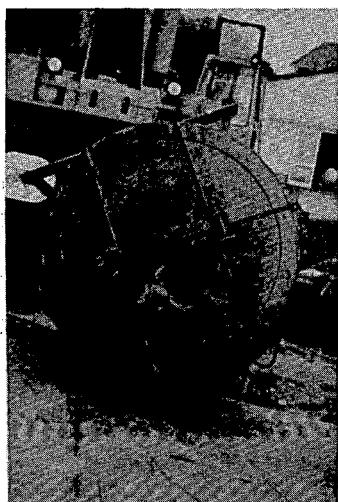


Figure 14. Overpack damage caused by the perforation tests against the package bottom (package n. 4563/ BTN 2094)

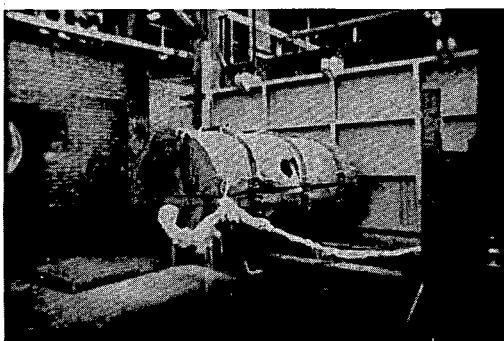


Figure 15. The package n. 4561/ BTN 2103
Before the furnace test

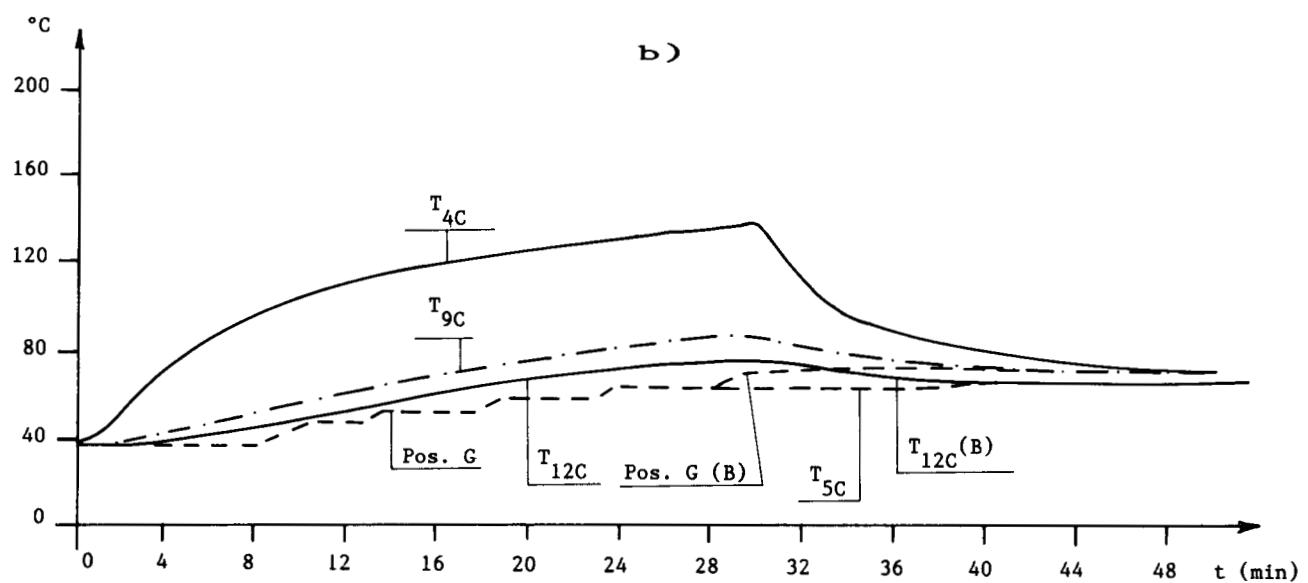
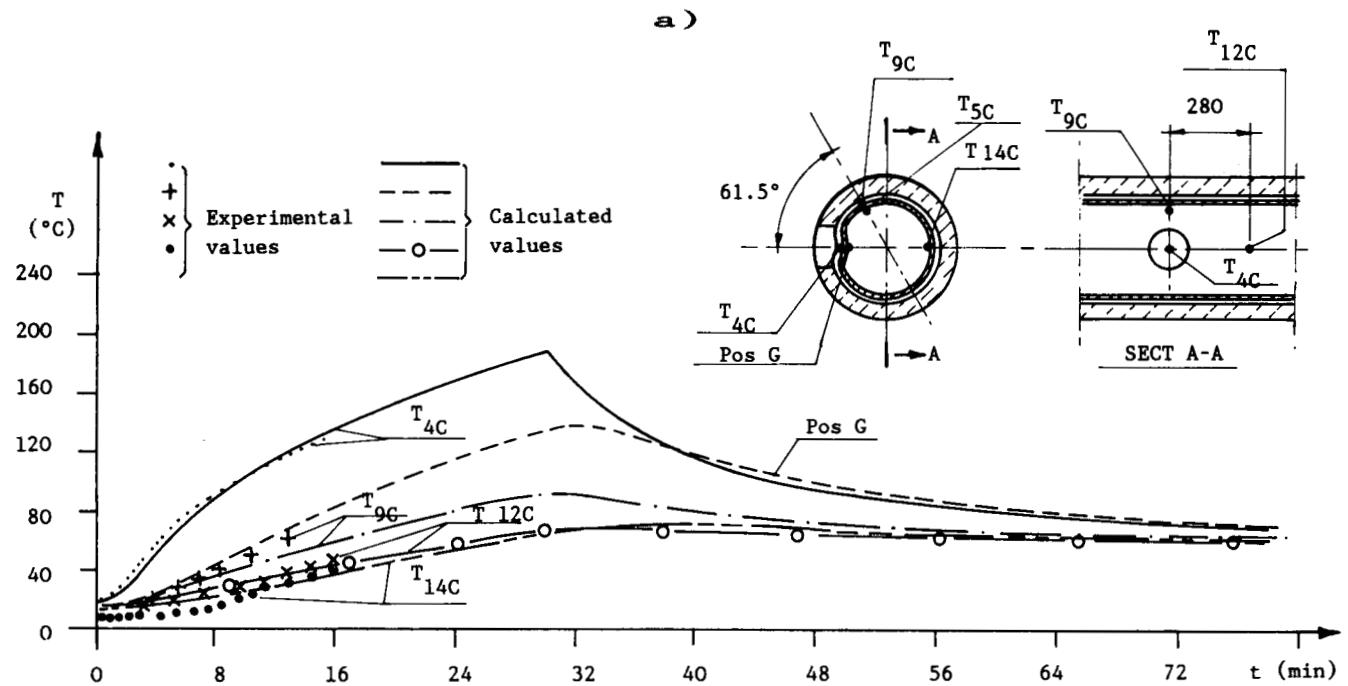


Figure 16. Package temperature versus time diagram:
 a) Comparisons between experimental and calculated packages temperatures
 b) Simulation of the UF6 real contents: 30B container temperature - time diagram

Test n.	package	Test sequence	Leakage Rate (<u>mbar.l</u>) s	air mass rate (<u>g/s</u>)	NOTE
1	4561	before the tests	$1.143 \cdot 10^{-2}$	$1.377 \cdot 10^{-5}$	
2	4561	after 9 mt free drop test and perforation test	$1.408 \cdot 10^{-2}$	$1.696 \cdot 10^{-5}$	
3	4561	After the furnace test	$4.46 \cdot 10^{-3}$	$5.373 \cdot 10^{-6}$	
4	4563	before the tests	$5.057 \cdot 10^{-2}$	$6.092 \cdot 10^{-5}$	
5	4563	After the 9mt corner free drop test and perforation test	19.8	$2.44 \cdot 10^{-2}$	plug valve screwed by hand
6	4563	"	3.038	$3.744 \cdot 10^{-3}$	plug valve screwed by tool
7	4563	before the 9mt lateral drop test	$2.8 \cdot 10^{-1}$	$3.374 \cdot 10^{-4}$	new valve
8	4563	After the 9mt lateral drop test and furnace test	$2.039 \cdot 10^{-1}$	$2.456 \cdot 10^{-4}$	

Tab. I : Results of the leaktightness tests

UPDATE ON PACKAGING FOR URANIUM HEXAFLUORIDE TRANSPORT

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ABSTRACT

The slightly enriched product UF_6 shipped from the enriching plants for the world's nuclear power plants must be protected in order to conform to domestic and international transport regulations. The principal overpack currently in use is the U. S. Department of Transportation (DOT) Specification 21PF-1 which protects Model 30 UF_6 cylinders (Title 49, Code of Federal Regulations; Part 178.121, Specification 21PF-1; Fire and Shock Resistant, Phenolic - Foam Insulated Overpack [Horizontal Loading]). Operational problems have developed due both to design and lack of maintenance, resulting in the entry of water into the insulation zone. Following major review of these problems, particularly those concerned with water entry and general deterioration, design modifications have been proposed. These modifications for existing overpacks are to be made only after any water absorbed within the phenolic foam insulation is reduced to an acceptable level. New overpacks will be fabricated under an enhanced design. Existing overpacks which are modified will be designated as 21PF-1A while new overpacks fabricated to the enhanced design will be designated as 21PF-1B. In both cases, proposed quality assurance/control requirements in the fabrication, modification, use and maintenance of the overpacks are applicable to fabricators, modifiers, owners and users. A composite report describing the proposal has been prepared.

INTRODUCTION

In order to meet the packaging requirements of domestic and International Atomic Energy Agency (IAEA) transport regulations, the cylinders of slightly enriched UF_6 shipped from the world's enriching plants are contained within overpacks. The 21PF-1 overpacks have been utilized to provide the required mechanical and thermal protection. A prototype which was subjected to the hypothetical accident test series in the mid-1960s, successfully provided the necessary protection. After being specified as a DOT specification package, it has been accepted for world-wide use. The current design of the 21PF-

1 overpack has features which require inspection and maintenance. These features include metal components fabricated of carbon steel and a non-metal covered closure interface of wood which steps down from outside to inside. When routine maintenance is not provided, the combination of warping of the wood in the step-joint and corrosion of the carbon steel permits water to enter the insulation cavity. The process is accelerated due to exposure to the moist, corrosive atmosphere during transport on sea-going vessels. In 1984, the U. S. Department of Energy (DOE) filed a petition for change to the DOT regulations to modify the design of the 21PF-1 overpacks. The Notice of Proposed Rulemaking was published as a Notice in the U. S. Federal Register. The modifications were to apply to existing overpacks as well as overpacks fabricated in the future. Since 1984, there has been additional testing to assure that the insulation in the existing overpacks after moisture removal provides the required protection. Subsequent design improvements have been submitted by the DOE to the DOT for the final rulemaking. The DOE is also proposing mandatory quality assurance/control requirements for the fabrication, modification, use, and maintenance of the 21PF-1 overpacks. A composite proposal was submitted to DOT by DOE in August 1987.¹ DOE recommended that the proposal be published in the Federal Register as a DOT final rule.

(This final rule had not been published at the time this paper was prepared; however, its publication is expected to be early in 1988.)

MODIFICATIONS TO U. S. DEPARTMENT OF TRANSPORTATION SPECIFICATION 21PF-1 OVERPACKS

The modifications of the 21PF-1 overpack design are basically to upgrade and enhance previously fabricated and new overpacks to provide regulatory protection to the contained cylinders of UF_6 and to minimize routine maintenance. In order to assure that the fabrication and modifications are made as specified and that the overpacks are maintained, the DOE is proposing mandatory quality assurance/control requirements which cover the fabrication, modification, use, and maintenance of the 21PF-1 overpacks. These requirements would apply to fabricators, modifiers, owners, and users. Thus, domestic fabricators and modifiers would be required to have a U. S. Nuclear Regulatory Commission (NRC) approved quality assurance program. Owners and users would also be required to have an NRC approved or equivalent program. (At the time this paper was published, DOT was considering

that for those overpacks which were fabricated and/or owned by non-U. S. firms, a quality assurance program which was approved by the appropriate IAEA competent authority would be required for overpacks transported within the U. S.). This is intended to assure that the 21PF-1 overpacks meet all prescribed regulatory standards. The design modifications are divided into two groups - for existing overpacks and for new overpacks.

The existing 21PF-1 overpacks must be thoroughly inspected prior to initiating modification and to assure that the moisture content of the phenolic foam insulation is sufficiently low. A drying procedure to remove absorbed water has been developed. Tests such as those described in the composite proposal of this paper verify that the dried phenolic foam provides an appropriate level of protection.

Proposed modifications to the existing 21PF-1 overpacks include:

1. Covering the lower step joint with carbon steel which is continuously welded to the inner and outer skins of the overpack. Painting step joint with intumescant paint. See Figure 1.
2. Installing two one-piece molded gaskets made of Silastic E RTV rubber.
3. Drilling holes in the longitudinal stiffener angles.
4. Providing specifications for welding and corrosion repair.
5. Sealing joints between stiffeners and outer shell.
6. Covering vent holes in outer shell with plastic plugs.
7. Detailed instructions for weighing.
8. These overpacks will be redesignated as "DOT Specification 21PF-1A."

Proposed modifications to 21PF-1 overpacks fabricated in the future include:

1. Changing wood materials from hard or sugar maple to white oak.
2. Changing metal parts from carbon steel to stainless steel, Type 304-L for sheet, plate and angle and flat bar and to 300 series for other parts.
3. Specifying welds as continuous, full penetration.
4. Reversing step joint with the step being upward from outside to inside and covering both upper and lower joints with steel. Painting step joint with intumescant paint. Step joint closure with metal-to-metal contact at outer step. See Figure 2.

5. Replacing gaskets by single one-piece molded gasket made of Silastic E RTV rubber.
6. Adding detailed instructions for weighing.
7. These overpacks will be designated as "DOT Specification 21PF-1B."

As part of the routine maintenance program, periodic inspections would be required. Each overpack would be inspected and recertified at intervals not exceeding five years. The date of recertification would be stamped on a new data plate. This recertification would include a determination that overpacks fully met the requirements of the DOT specification and the applicable engineering drawings, that necessary repairs had been made and the packaging tare weight remained within a 25-pound limit. Details for weighing are specified.

CONCLUSION

The DOT specification overpack has been the "work horse" for transport of slightly enriched UF₆. Design deficiencies and lack of maintenance have resulted in the entry of water into the insulation, deterioration of the overpacks, and created problems in transport. The proposed design modifications and additional requirements for quality assurance/control and maintenance should enhance the safe transport of slightly enriched UF₆ and greatly extend the useful lives of the DOT Specification 21PF-1 overpacks.

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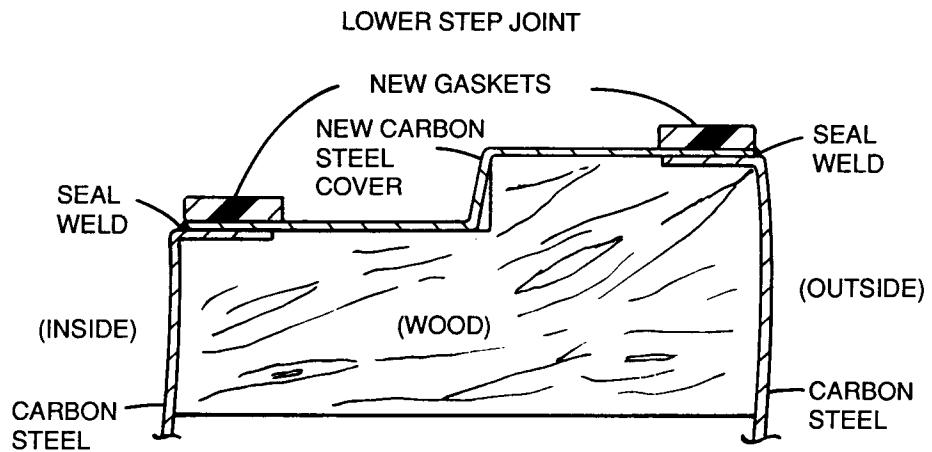


Fig. 1. Proposed Modifications to Existing D.O.T. 21PF-1 Overpacks
Designated "DOT 21PF-1A"

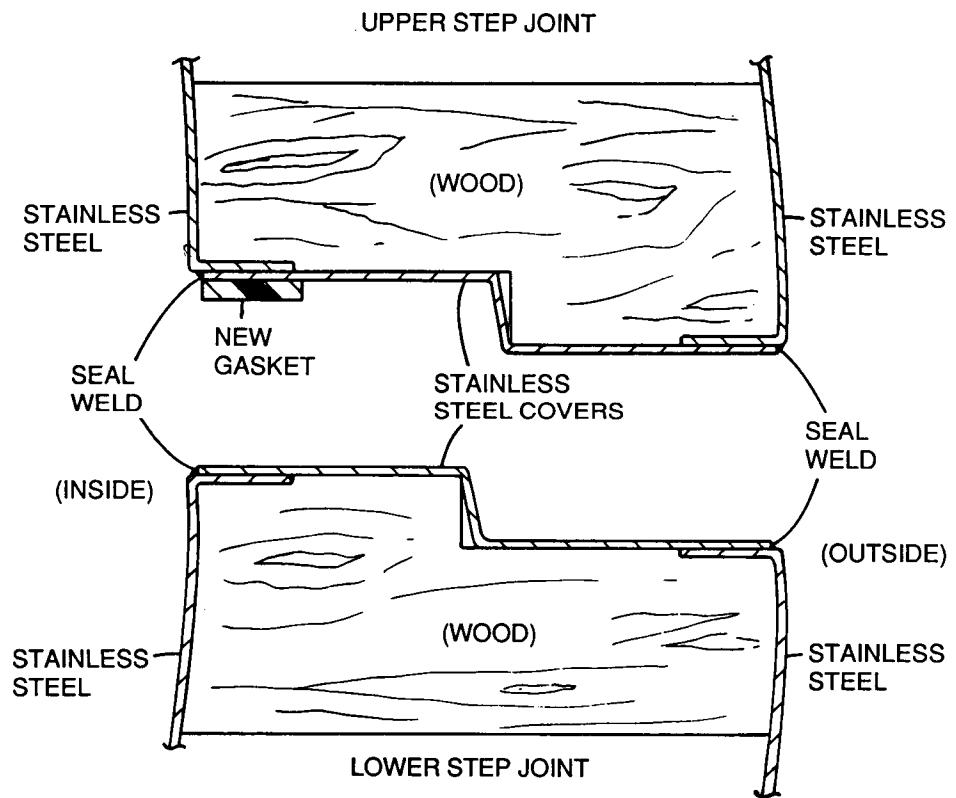


Fig. 2. Proposed Modifications to New D.O.T. 21PF-1 Overpacks
Designated "DOT 21PF-1B"

THERMAL PROPERTIES EVALUATION OF UF₆ CYLINDER OVERPACK INSULATION

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ABSTRACT

A phenolic foam is incorporated into the design and construction of UF₆ cylinder shipping containers known as overpacks. The overpack provides mechanical and thermal protection to UF₆ product cylinders during transport. The phenolic foam is utilized for thermal insulation. An important criterion in overpack design is to limit temperature excursions of the product cylinder below the triple point of UF₆, preventing possible overpressurization of the cylinder. A current requirement is that the product cylinder wall temperature be maintained below the UF₆ triple point with the outer surfaces of the overpack exposed to 1475°F for thirty minutes. A possible heat source is the close proximity of a fire. Overpack thermal protection design requires thermal properties data for the materials of construction, particularly for the phenolic foam insulation which provides the primary thermal resistance. Thermal conductivity and heat capacity data for the phenolic foam are limited in the literature. Accurate heat transfer models of the overpack require thermal properties spanning the range from room temperature to 1475°F. An experimental program was devised to measure the thermal conductivity and heat capacity of the insulating foam over the temperature span of interest.

TEXT

Enriched uranium in the form of solid uranium hexafluoride is transported in interstate and international shipments in cylinders which are enclosed within an insulated, protective overpack. Protective overpacks have been in service for a number of years with the intervening introduction of minor design modifications. Environmental and safety requirements have dictated additional testing, analysis, and evaluation of overpack design and construction related to possible accident scenarios. The phenolic foam insulating material which is encased within a wood reinforced shell assembly forming the overpack is an integral part of the uranium hexafluoride cylinder protection. The insulation barrier provides a measure of mechanical shock isolation in addition to its primary function of serving as a thermal resistance to possible heat sources to which a product cylinder might be exposed during transport. Properties of the insulating foam are necessary in order to determine both mechanical and thermal functions of the

material. Thermal properties of the insulation are necessary for incorporation into heat transfer analyses of the overpack subjected to known or assumed thermal loads. The properties of solid uranium hexafluoride dictate design requirements for the overpack in terms of thermal protection; reasonable means must be provided in order to prevent the triple point temperature of the product from being attained. Should the triple point temperature be reached, overpressurization of the containment cylinder could occur with the possibility of a breach with an accompanying environmental insult. The phenolic foam is required to function over a temperature range of approximately 1400°F; a temperature range which could be produced by a close proximity fire. The insulating foam is arranged in an annular ring around the product cylinder with end closures. Temperatures in the insulation will vary with radial location during overpack exposure to a transient thermal excursion; therefore, it is important to determine thermal properties of the insulation at various temperatures over the range of interest. The determination of the temperature correlation of thermal properties will enhance analytical thermal models which may be developed.

EXPERIMENTAL PROCEDURE

Thermal properties of interest to the development of analytical heat transfer models are thermal conductivity and specific heat capacity. The thermal conductivity of the overpack phenolic foam was measured using an apparatus conforming to the requirements of the American Society for Testing and Materials (ASTM), Standard C177, Standard Method of Test for Thermal Conductivity of Materials by Means of the Guarded Hot Plate. This test method is recommended for insulating materials over a large range of temperatures for the condition of steady state heat flow. The method consists of generating axial heat flow through two samples of the material to be tested, placed on either side of a known heat source. Auxiliary heat sources provide isothermal sample cold surface temperatures while additional controlled heaters and insulation reduce radial heat losses from the test stack assembly to acceptable levels. The method consists of achieving steady state conditions in the test assembly, measuring the temperature gradient across a known sample thickness and determining the heat flow generated in the test stack assembly. The thermal conductivity of a homogeneous material is defined as the time rate of heat flow through unit area per unit temperature

gradient in the direction perpendicular to an isothermal surface. Eight-inch diameter, one-half inch thick disk samples of the phenolic foam were tested in a nitrogen atmosphere at mean sample temperatures to 1285°F.

The specific heat capacity of the phenolic foam overpack insulation was determined by means of the method of "drop calorimetry". The heat required to raise the temperature of a unit mass of material by one degree is the heat capacity at constant pressure. The specific heat capacity is expressed in terms of calories per gram per °F.

The drop method can be utilized to measure heat capacity or heats of fusion of transition. A sample is dropped from a hot furnace at a known elevated temperature into a calorimeter maintained at some known, lower temperature. The heat absorbed by the calorimeter is equal to the difference in heat content between the initial sample temperatures and the final sample temperature. The configuration of the calorimeter was such that the device functioned as a isothermal water calorimeter; whereby, the calorimeter proper is surrounded by a constant temperature jacket. The calorimeter consisted of a dewar flask equipped with electric stirrers to maintain a constant 25°C water bath temperature. The dewar vessel was surrounded by foam insulation in order to prevent heat exchange from the outer dewar wall to atmosphere. An elevated temperature furnace was mounted on supports above the calorimeter bath, and the furnace was capable of obtaining temperatures of ~1800°F. A calorimetry experiment consisted of elevating the temperature of the sample to 1475°F, dropping the sample into the calorimeter, and measuring the temperature increase of the known water mass as a function of time. The specific heat capacity of the phenolic foam could be then computed from the measured temperature rise of the calorimeter fluid.

RESULTS

The thermal conductivity of the phenolic foam increases with increasing temperature, exhibiting an approximately linear relationship to 1000°F. A non-steady state value at mean sample temperature of

1285°F revealed a significantly higher thermal conductivity and a deviation from the linear temperature thermal conductivity relationship obtained from room temperature to 1000°F. The higher thermal conductivity at temperatures exceeding 1000°F is attributable to compositional changes in the material and to an increase in the relative contribution from the mechanism of radiative heat transfer at the higher temperatures. Measured thermal conductivities ranged from 0.020 Btu·ft/h·ft²·°F at 145°F to 0.070 btu·ft/h·ft²·°F at 1000°F. The transient test at 1285°F indicated a thermal conductivity of 0.154 Btu·ft/h·ft²·°F.

The specific heat capacity for the phenolic foam at an initial temperature of 1475°F for five samples tested ranged from 0.093 calories/gm/°F to 0.106 calories/gm/°F. The experimentally measured thermal properties of the phenolic foam are presented for incorporation into heat transfer models of the overpack container.

FOLLOW ON ACTIVITIES

Thermal Conductivity

1. Additional testing to determine statistical variance.
2. Determine effects of material decomposition on thermal conductivity and structural integrity, particularly at temperatures exceeding 1000°F. Time at temperature in an oxidizing atmosphere is postulated as being a primary factor related to decomposition of the phenolic foam insulation.
3. Determine the effects of material anisotropy on thermal conductivity.

Specific Heat Capacity

1. Refine measurement technique and utilize other methods for comparison.
2. Determine specific heat temperature correlations for the phenolic foam material.

Prepared by the Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tennessee 37831, operated by MARTIN MARIETTA ENERGY SYSTEMS, INCORPORATED, for the U.S. DEPARTMENT OF ENERGY, under Contract No. DE-AC05-84OR21400.

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INVESTIGATION OF THE THERMAL BEHAVIOR OF 2 1/2 TON CYLINDER PROTECTIVE OVERPACK

by
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ABSTRACT:

UF₆ cylinders containing reactor grade enriched uranium are transported in protective overpacks. Recently, the design of the 2 1/2 ton UF₆ cylinder overpack was modified to insure the safety of the cylinder inside the overpack. Modifications include a continuous stainless steel liner from the outer surface to the inner surface of the overpack and step joints between the upper and lower halves of the overpack.

The effects of a continuous stainless steel liner and moisture in the insulation layer of a UF₆ cylinder protective overpack were investigated with a numerical code. Results were compared with limited available field data. The purpose of comparing the numerical results with field data is to insure the validity of the numerical analysis and the physical properties used in the analysis. The study indicates that the continuous stainless steel liner did not influence the heat transfer rate much from the outer surface of the overpack to the 30B cylinder inside. The effect of step joints was not modeled due to the difficulty of quantifying the leakage rate through the gap. With a continuous stainless steel liner from the outside of the overpack to the inside, the overpack satisfies the thermal design criteria of protecting the cylinder inside for a minimum of 30 minutes when the overpack is exposed to a fire. The effect of moisture inside the insulation layer in the overpack is to reduce the energy to the cylinder with its high thermal capacity. The high pressure steam generated from the moisture will be relieved externally through the vent holes on the outer surface of the overpack. Although these holes are sealed after the overpack is dried, the plug sealing the holes will melt when the overpack is exposed to a fire.

INTRODUCTION

A majority of the commercial nuclear reactors utilize uranium, enriched as uranium hexafluoride (UF₆) to an assay of 3.0%, for fuel. The UF₆ is transported throughout the country in 30-inch diameter cylinders which are encased in protective overpacks. The function of the overpacks is to prevent release of UF₆ by protecting the cylinders from physical damage and providing thermal insulation in case of fire. A cylinder will rupture when the internal pressure exceeds the ultimate hoop stress of the cylinder. Internal pressure, sufficient to rupture a cylinder, can be developed by expansion of liquid UF₆ on heating if the cylinder is completely filled with liquid UF₆. The thermal design criteria specifies that the overpack must protect the cylinder at least one-half hour in an oil fire. The purpose of this study is to verify compliance of the thermal design criteria with design modifications. These include the step joints between the upper and lower halves of the overpack and the use of a continuous stainless steel liner from the outer to the inner surface of the overpack. The study also includes the effect of moisture absorbed by the insulation layer of the overpack. The study was conducted with a numerical program, TRUMP¹ developed by Lawrence Livermore National Laboratory. A two-dimensional model of a cylinder and an overpack is used for the investigation. The exact nature of heat flux from a fire is a very complicated phenomenon involving the nature of fuel (gas or liquid), combustion conditions, and many other factors. Because of the complexity of the heat flux and the purpose of the study in determining the effects of geometrical variations of the overpack to the safety of the cylinder, the source heat flux was simplified and held constant for all cases.

THERMAL ANALYSIS MODEL

TRUMP is a general purpose, heat transfer computer code capable of handling multidimensional systems with conduction, convection, and radiation heat transfer processes. TRUMP solves sets of nonlinear parabolic partial differential equations for both steady and transient cases.

The overpack for a 2 1/2 ton UF₆ cylinder is a horizontal loading type having two halves as shown

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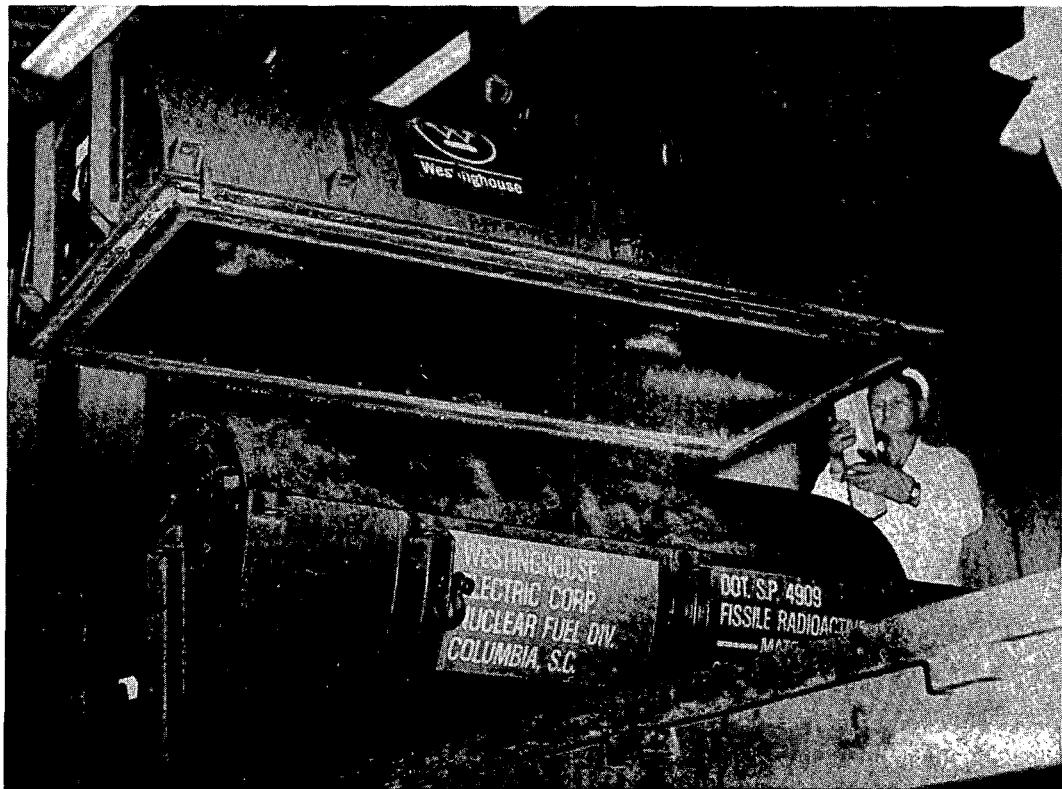


Fig. 1. 2-1/2 ton UF_6 cylinder overpack - horizontal loading type.

in Fig. 1. A two-dimensional model, shown in Fig. 2, was used to investigate the temperature profiles of a prototype cylinder in cases where flames surrounded the overpack. The solid UF_6 mass inside a cylinder was assumed to be deposited radially in uniform thickness layers inward from the cylinder wall to simplify the analysis. In the case of a full cylinder, solid UF_6 occupies more than 60% of the internal surface of a cylinder, which makes this simplification close to the real situation. The heat flux from a fire to an overpack is the sum of the radiative and convective heat transfer processes, and is expressed as

$$q = A * [F_{12} * (T_f^4 - T_s^4) + h_c (T_f - T_s)]$$

where F_{12} is the overall exchange factor, h_c is the natural convective heat transfer coefficient, and T_f and T_s are the temperatures of the fire and the surface of the overpack, respectively. The cylinder surface temperature, T_c , will be the main observation point in the analysis. The heat exchange between the inner surface of an overpack and the outer surface of the cylinder is by radiation only. The initial temperature of the overpack system is 80°F. To simulate exposure to a fire, the overpack was suddenly exposed to a gas environment with an average temperature of 1,750°F. The gas emissivity used was 0.5. The effect of moisture in the insulating layer was analyzed by assuming that the total moisture content is concentrated in the middle of the insulation layer

as shown in Fig. 3. Since the primary purpose of the investigation was to determine the temperatures of the cylinder and UF_6 , the assumption did not introduce any gross error into the final analysis. For analysis of the worst case, an overpack is assumed to be engulfed in fire.

RESULTS

The effectiveness of the overpack in meeting the thermal design criteria was investigated with the two-dimensional model. The worst case considered was an infinitely large fire source with the flames surrounding the entire overpack. The results of this case are compared in Fig. 4 with experimental data obtained by the Oak Ridge Gaseous Diffusion Plant (ORGDP) in 1966². The agreement between the analytical and experimental results is fairly good considering the uncertainties that exist in experimental conditions. The cylinder surface temperature at the mid-section by the analysis is approximately 122°F after one hour in the fire even though the outside surface of the overpack is at 1,742°F. Furthermore, the UF_6 temperature inside the cylinder is below the triple point temperature. Even if the higher values of the experimental data are chosen, the cylinder surface temperature after one-half hour in the fire would be below the triple point temperature of UF_6 . This indicates that the overpack meets the thermal design criteria of protecting the cylinder for one-half hour when exposed to a fire.

Fig. 3. Model of cylinder and protective overpack with moisture.

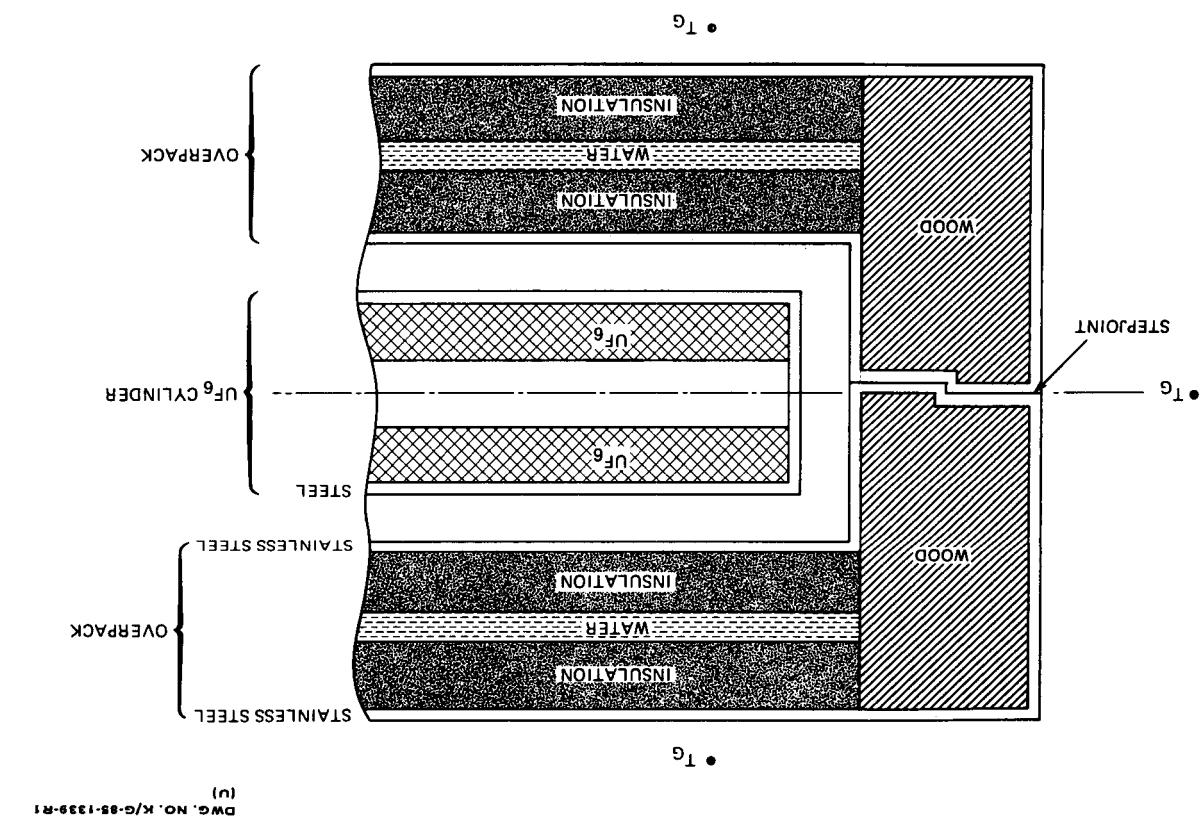
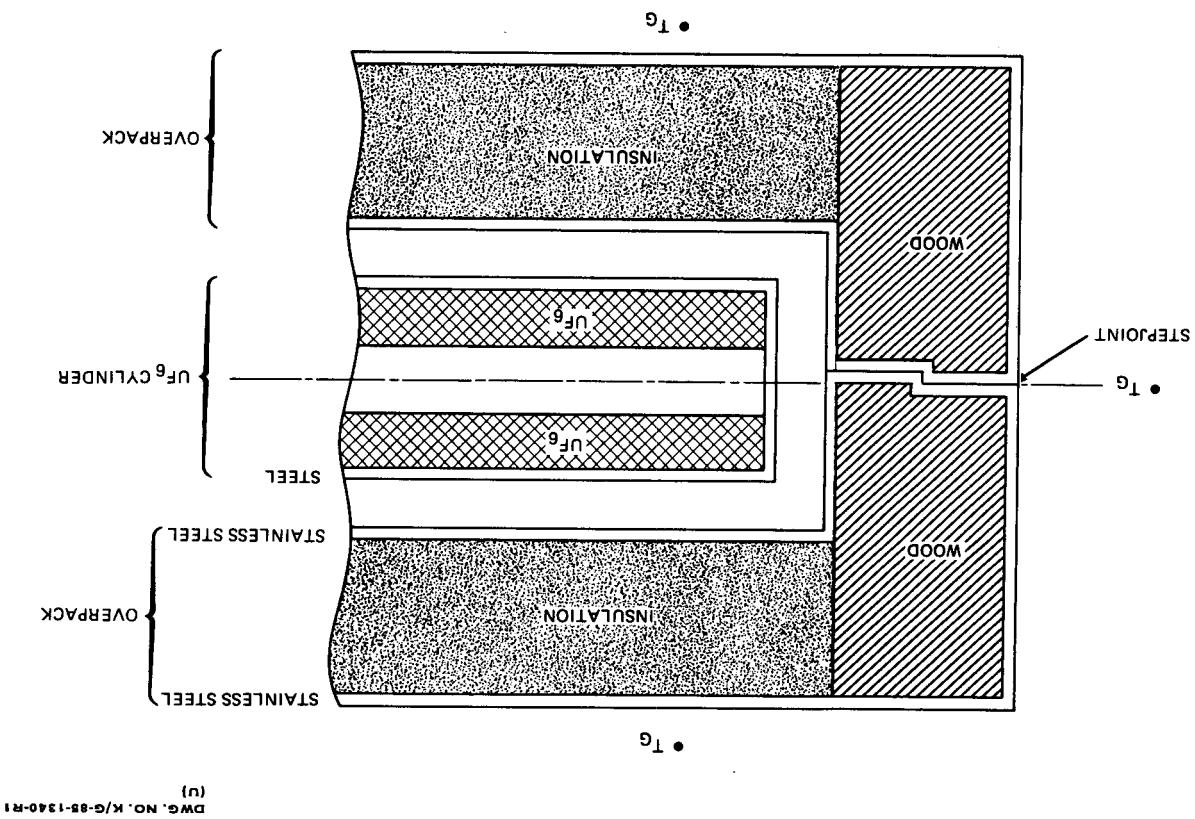


Fig. 2. Model of cylinder and protective overpack.



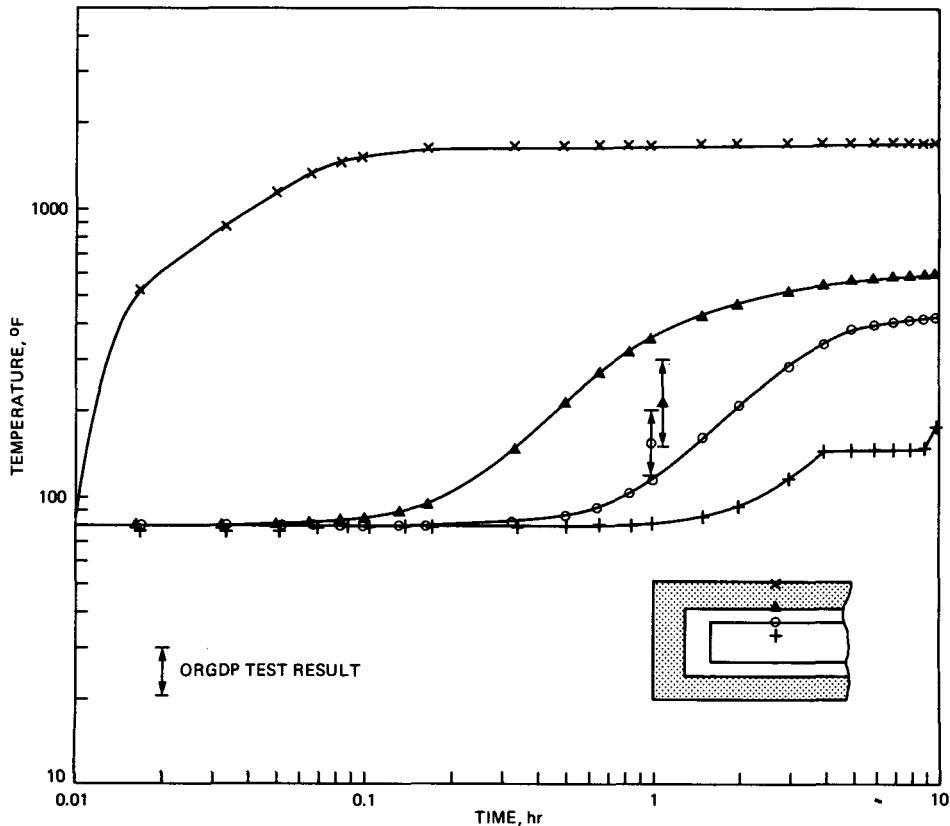


Fig. 4. Radial temperature variation at mid-section of the overpack with time in fire.

The phenolic foam used in overpacks was manufactured based on some physical specifications such as density, porosity, etc., but no thermal property specifications. Since there was no record of a measured thermal conductivity value, a conductivity value that best fit the existing test results was obtained. Hence, a thermal conductivity value of 0.170 Btu/hr ft°F was used in this analysis, which was the best fit to the existing data². The validity of the code and other physical properties used in the code are proven in Fig. 5, where the temperature profile through the ends, whose thermal conductivity value is known, agreed well with the experimental value. The thermal conductivity value reported by Frazier, however, was one order of magnitude lower than the value used in the analysis at room temperature, but was comparable at temperatures above 1,000°F. The temperature profile of the overpack with temperature dependent thermal conductivity by Frazier was plotted in Fig. 6 along with the constant thermal conductivity of 0.170 Btu/hr ft°F and ORGDP experimental value. The overpack temperature profile with the constant thermal conductivity case in the cylinder safety study. The temperature profiles with both thermal conductivity values satisfy the thermal design criteria.

The effectiveness of the overpack step joint in limiting the cylinder wall temperature rise will depend entirely upon the leakage rate of hot flume through the gap which is difficult to quantify.

Theoretically the step joint will be more effective than the straight joint in terms of reducing the leakage rate of hot flume from the fire. In addition, the step joints between the two halves of the overpack would provide a more solid connection structurally. Therefore, there is no quantitative criteria of leakage rate through the gap in introducing the step joint. Thermal analysis of the step joint was not performed. The effect of a continuous stainless steel liner from the outer to the inner surface of the overpack was examined, but this effect is minimal, as shown in Fig. 7. The liner is 14 gauge stainless steel, and the heat transfer rate through this cross section is small compared to that through the insulation layer.

The effect of moisture in the insulation layer of the overpack was investigated with the model shown in Fig. 3. There are 40 vent holes on the outer surface of overpack. During the overpack construction process, a fully assembled overpack is dried in an oven to drive away any moisture contained in the insulation layer of the overpack. When an overpack is dried, the vent holes are covered with a plastic plug to make it moisture proof. But the insulation layer of an overpack absorbs moisture during long years of service, thereby changing the thermal properties of the insulation material. In this analysis, a total of 200 pounds of moisture was assumed to be absorbed by the insulation layer, even though the amount of moisture absorbed by the phenolic foam varied widely. The 200 pounds used

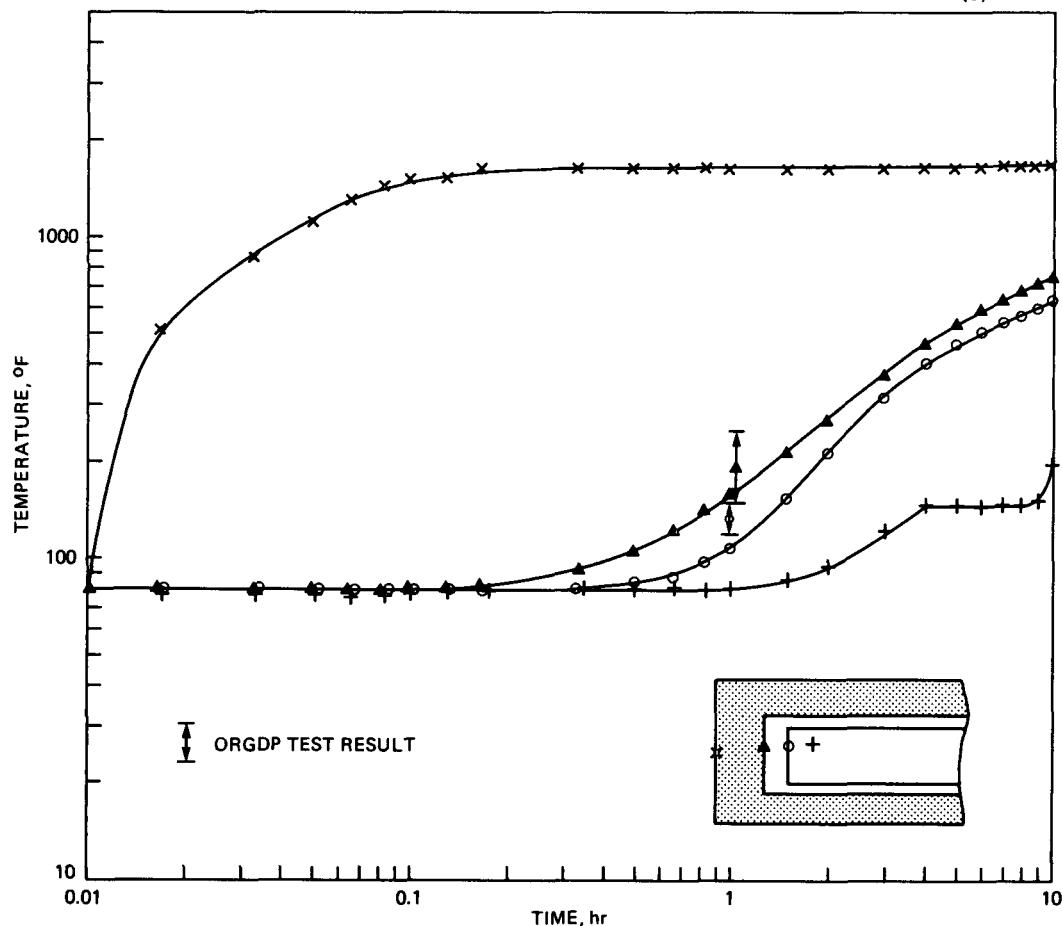


Fig. 5. Temperature variation with time--end section.

in this analysis is based on actual measurement of overpack weight at saturation condition. The effect of moisture in the insulation layer is to reduce the energy to the cylinder inside because the heat capacity of water is greater than the insulation layer and because of the latent heat of the moisture. This is evident in Fig. 8, where the radial temperature profiles from the outer surface of the overpack to UF_6 are plotted with and without moisture in the insulation layer of the overpack. The result indicates that the temperature of the inner surface of an overpack decreased by 50% with 200 pounds of moisture, and the average water temperature was still below its boiling point. Any danger of high pressure steam developed from a long exposure in a fire condensing on the UF_6 cylinder is remote due to the fact that the plugs sealing the vent holes on the outer surface of the overpack will melt before such pressure would develop in the insulation layer. Even if such pressure would develop in the insulation layer, such pressure would be more critical to the outer surface of the overpack than the inner surface due to the combination of longer radius of curvature and higher degradation of strength of the outer surface at the elevated temperature.

CONCLUSIONS

The cylinder with overpack which is used to transport 3.0% U^{235} enriched uranium has been investigated. The study proves that the overpack with a step joint and continuous stainless steel liner is capable of protecting the cylinder for at least half an hour in a fire at an average temperature of 1,750°F. Moisture inside the insulation layer acts as a heat sink to reduce the heat flux to the cylinder. There seems to be no danger from any high pressure steam developed in the insulation layer. This may be due to the fact that the plastic plugs covering the vent holes on the outer surface of the overpack will melt and relieve pressure. The step joint will reduce hot flume leakage while increasing the tightness at the joints. Actual cases may differ from this simple analysis due to variation of heat flux from fire and overpack physical condition. This analysis represents a worse than real case with fire surrounding the overpack and with a constant thermal conductivity value of the insulation at all temperature ranges. The overpack, even in these situations, satisfies the thermal design criteria of protecting the cylinder for more than one-half hour when exposed to a fire.

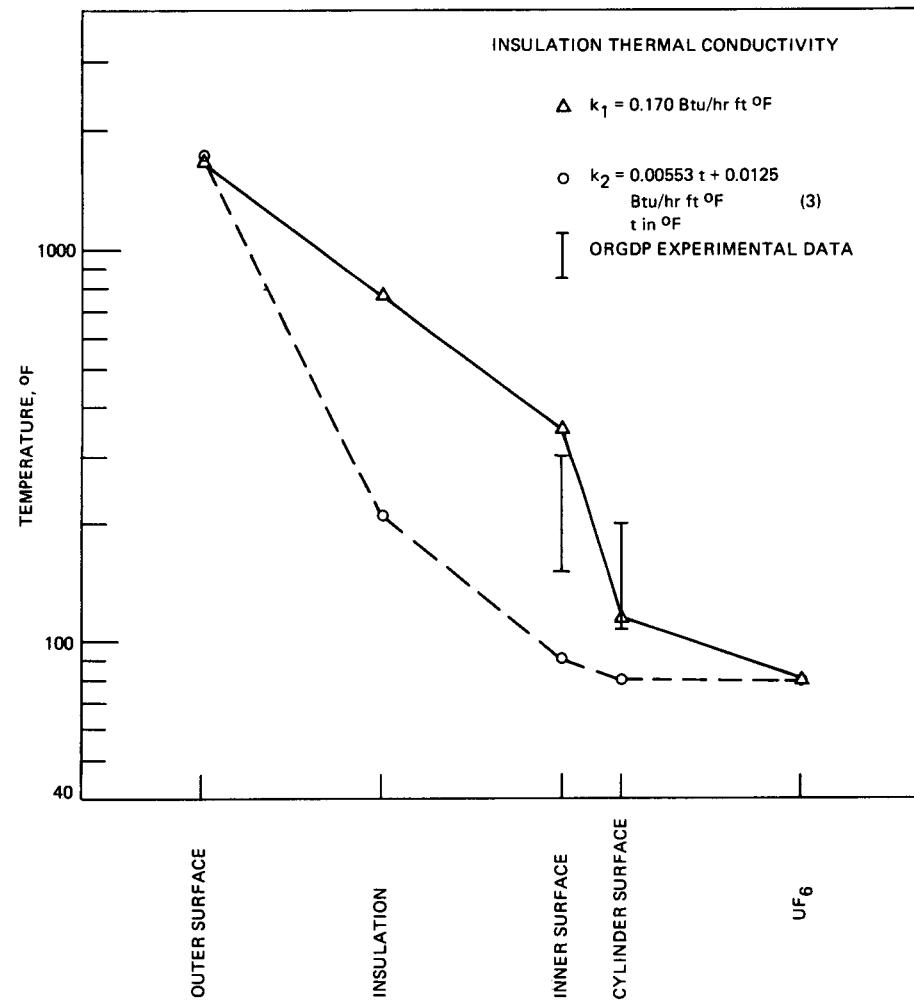


Fig. 6. Variation of temperature profile with thermal conductivity of insulating material.

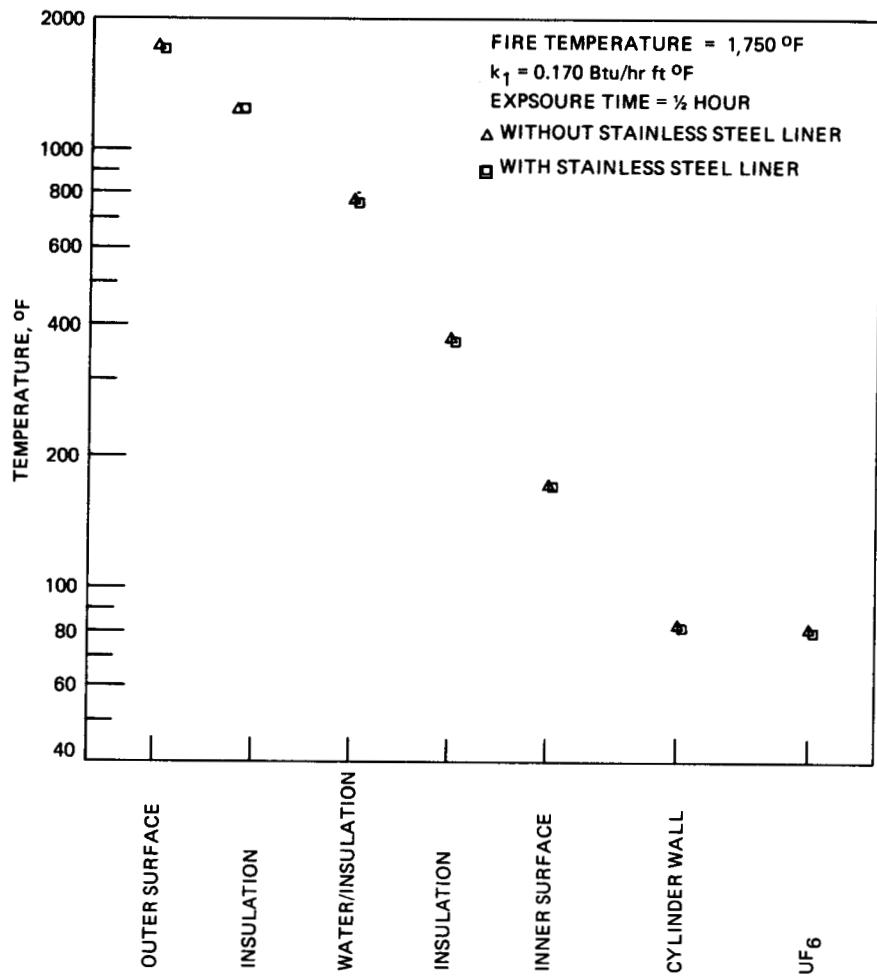


Fig. 7. Effect of continuous stainless steel liner on overpack temperature profile.

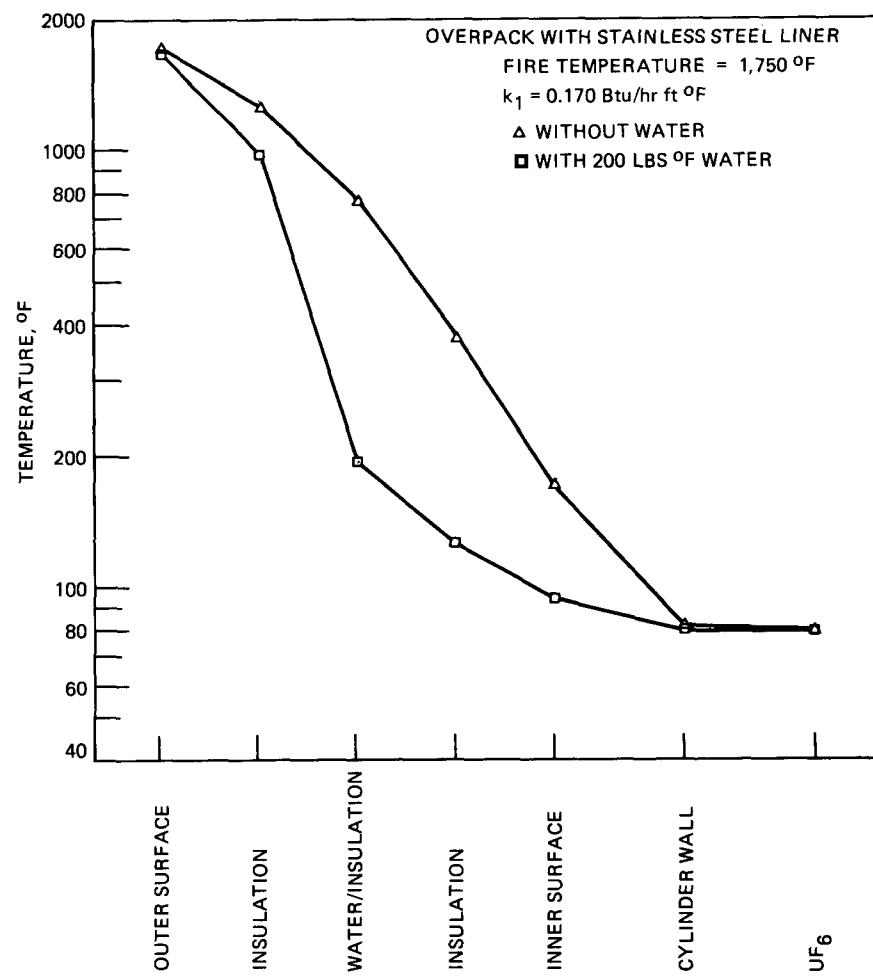


Fig. 8. Effect of moisture in the insulation layer on overpack temperature profile.

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ONE-INCH UF₆ CYLINDER VALVE FAILURE

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ABSTRACT

Defects in one-inch cylinder valves procured for use in uranium hexafluoride (UF₆) cylinders were discovered during installation.

The defects were detected by soap testing of the valves and cylinders. The investigation detected similar problems in other production runs by the same manufacturer. This paper covers the investigation of the original problem, evaluation of the defects, notification of all concerned parties, interim solution, and corrective actions taken to eliminate this problem.

INTRODUCTION

The Paducah Plant, Martin Marietta Energy Systems, Inc., procures one-inch angle valves for installation in cylinders that are used for storage and transportation of uranium hexafluoride (UF₆). In June 1986, a contract was awarded to the Superior Valve Co., Washington, Pennsylvania, for 2694 valves. After delivery had started in March 1987, the Paducah Plant began to forward valves to the fabricator of new tails cylinders and to the Portsmouth Plant for normal inventory requirements. In July, the fabricator discovered that two valves from forging heat lot 20 were leaking after installation into cylinder couplings.

INVESTIGATION

The materials laboratory at the Paducah plant began an investigation of the cracked valves. When five additional fractures, from forging Heats 17 and 20, were discovered, the manufacturer was informed of the incidents; and the Portsmouth Plant and cylinder fabricator were notified that valves from Heats 17 and 20 were not to be used. The Department of Energy (DOE) was notified of the failures, and they subsequently contacted the

Nuclear Regulatory Commission (NRC) so that their licensees would be aware of the problem. The scope of the investigation was expanded to include assistance from other diffusion plants and Oak Ridge National Laboratory (ORNL). The valve manufacturer and the forging plant also initiated independent investigations.

The Paducah Plant investigated the reliability of other forging lots by installing valves from Heats 18, 19, and 21 into cylinders and leak testing with 100 psi air. After a valve from Heat 19 broke off during installation, all valves from Heats 17 through 22 were considered suspect; and a hold was put on their use. The valves could not be put into a cylinder, and any cylinder with a suspect valve could not be filled or fed until the valve was changed. Initially, the suspect group included valves from Heat 16 because one from that group had been included in the 1987 delivery. However, it was determined that Heat 16 forgings had been produced at an earlier date and that no problems had been encountered with valves machined from production related forgings.

Preliminary information from the Paducah laboratory and ORNL indicated that high concentrations of aluminum were present on fracture faces. It was suspected that segregation of aluminum could have occurred before the bodies were forged. Visits to the bronze mill, forging plant, and valve manufacturer were planned to discuss preliminary findings, observe manufacturing methods, and review quality assurance programs.

SHORT-TERM SOLUTION

A valve refurbishment program was initiated at the Paducah and Portsmouth Plants to relieve the shortage created by the hold put on the use of Heats 17 to 22. Uncontaminated valves that had been removed from clean cylinders, or decontaminated valves removed from rehydrogenated cylinders, were rebuilt to an approved specification and tested for acceptance. Valves rebuilt from uncontaminated parts are being forwarded to a cylinder manufacturer, while the use of valves rebuilt from decontaminated parts is limited to the enrichment complex. The use of rebuilt valves prevented any disruption to productive activities and allowed delivery of new cylinders.

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FAILURE ANALYSIS OF CRACKED VALVES

The discovery of cracks in the bodies of several one-inch cylinder valves (Figure 1) was a significant concern. The holds put on both the use of the suspect valves and the manufacture of new ones could not be removed until there was an understanding of the cause of the problem and corrective measures identified. The assistance of laboratory groups from the three diffusion plants and ORNL were employed in the investigation. As the studies progressed, visits were made to the valve manufacturer and his suppliers to determine how the various manufacturing steps could contribute to the cracking problem.

Laboratory Procedures

The investigation involved several analytical and testing facilities within the laboratories of Martin Marietta Energy Systems, Inc. The following laboratory investigations and methods were employed.

Valve bodies, cracked and uncracked from the suspect lots, and bodies from previous production lots were chemically analyzed to determine their conformance to specification limits.

Metallographic cross sections were prepared of numerous valves from both the suspect and previous production lots.

The tensile strength of both cracked and uncracked bodies was measured by loading through the inlet port pipe thread with a coupling attached to a rod and through the upper portion of the body with a stem brazed to a rod. Small tensile specimens were also machined from valve bodies.

The failure mode was investigated by examination of fracture surfaces in a scanning electron microscope.

The homogeneity of the metallurgical structure of the bodies was investigated by electron microprobe measurements and Auger spectroscopy.

The physical soundness of suspect bodies was investigated by use of penetrant methods, eddy current inspection, and radiography.

Experimental Findings

Spectrochemical analysis of 30 bodies from the suspect lots indicated that the composition of both cracked and uncracked bodies was within the specified limits for Alloy C63600. The alloy specification requires 3.0 to 4.0 Al, 0.7 - 1.3 Si, <0.35 Zn, <0.2 Sn, <0.10 Fe, <0.15 Ni, <0.15 As, and <0.03 Pb. Additional analysis by atomic absorption method revealed that the lead concentration was less than 0.01 wt %. The findings are consistent with data obtained from mill certification reports and tests that were conducted when the values were originally inspected.

The microstructures of both cracked and uncracked valves were single phase with an abundance of twins. However, the microstructure adjacent to the crack path (Figure 2) contained grain boundary

networks of mixtures of oxide and metallic phases (Figure 3).

Cracked valves pulled in tension failed in a ductile manner. The bodies yield at approximately 25,000 psi based on net area. Tensile specimens machined from the bodies of uncracked valves yielded at 20,000 to 22,000 psi. Similar specimens from extruded stock yielded at 43,000 psi.

Examination of fracture surfaces in the scanning electron microscope indicated that the service cracks were intergranular (Figure 4a). The surfaces of fresh fractures generated by laboratory testing were characteristic of ductile tearing (Figure 4b). Both failure modes were present on the fracture face of a leaking valve that had been pulled to failure.

Chemical analyses on microscopic structural features were performed with an electron microprobe. Analyses included traverses across the grain interiors and the grain boundaries as well as elemental maps of the grain structure. In the general single-phase microstructure, no evidence of segregation to the grain boundaries or significant compositional variations within the grains were detected. Analyses in the vicinity of cracks showed a different result. High concentrations of aluminum and oxygen were found to be associated with the films observed by optical microscopy.

In samples showing large, thick, film-like areas in the vicinity of cracks and exhibiting well-defined, separate phases (dark and light) within the film, microprobe results indicated that the dark phase, which was usually on the outside of the film, was rich in aluminum and oxygen while the interior, lighter phase was rich in copper and oxygen (Figures 5a and 5b).

The aluminum-rich oxide film (dark phase) often is found to completely enclose small islands of either light-grey oxide, which is rich in copper, or metal matrix or combinations of the two. Traverses away from the film-like region showed no signs of extensive aluminum depletion in the matrix.

Detailed Auger spectroscopy was performed on a fracture surface of a valve that cracked during installation and was subsequently tested to failure in a tensile test. The major portion of the entire fracture surface showed ductile failure which resulted from the tensile testing (Figure 6). Within the brittle fracture region, two different areas were identified. A portion of the brittle region appeared dark, and it is assumed this area corresponds to a crack in the valve that formed during or prior to installation and was present before the valve was broken during tensile testing. Lighter, brittle fracture areas were also observed; and presumably these areas correspond to "fresh", brittle fracture regions produced during tensile testing to failure to the specimen.

The aluminum concentration profile as a function of sputtering time is presented in Figure 7. Assuming the sputtering rate was constant at 1 nm/min throughout the analysis, the sputtering

time is linearly related to the depth of sputtering; and therefore a distance scale is superimposed on the time axis. Analyses from the dark, brittle area are marked B1 and B2. A third area on the dark surface that had a different morphology and was analyzed separately is marked B3. The results from the "fresh", brittle area are marked B4. Results from the ductile area are marked D1. The ductile area (D1) contained less than 10% aluminum, which is in good agreement with the level expected in the matrix. The brittle areas, B1, B2, and B4, contain quite high levels of aluminum, 40% to 70%, indicating significant aluminum enrichment. Finally, the area marked B3 was nearly pure aluminum (>90%). The remainder of the metallic constituent was copper.

The failures of the one-inch cylinder valve bodies prompted investigations to determine if non-destructive methods can be used to detect flaws in each of the material forms. Investigation by an ultrasonic method indicated that small, <1/8 inch, flaws could be detected within the forged body. Examination of the machined valve bodies indicated that surface flaws could be detected by a penetrant method if the solder coating on the threads of the inlet part was removed and the valve surface was chemically etched to remove disturbed metal. The use of eddy current methods for inspection of the bore of the inlet part was investigated. It was determined that the method would only detect through cracks or flaws that were within 20 mils of the bore surface.

The Metals and Ceramics Division at ORNL investigated the feasibility of locating flaws in the wall of the inlet port by the use of a special X-ray tube which produces a 360° panoramic exposure of the bore with a single radiograph. Indications of small ($\leq 1/8$ inch) flaws were observed in examination of the suspect valves. The utilization of the method is limited because the tin solder coating must be removed before inspection.

CONCLUSIONS

Intergranular networks of oxides were found in the vicinity of the cracks in the valve bodies. An isolated region of nearly pure aluminum was also identified on a fracture surface. This indicated that the original melt was probably inhomogeneous. Such nearly pure aluminum inhomogeneities could result in cracking during processing with subsequent heavy oxide formation along the cracks, or these areas could promote oxide formation which rendered the material susceptible to cracking. The inhomogeneity appeared to be localized within small volume elements of a few valves. It was not detectable by routine chemical or mechanical tests of the forging stock or finished bodies. The unflawed portion of a cracked valve body exhibited a ductile failure mode, and its microstructure was free of grain boundary films.

The Superior one-inch angel valve designated Heats 17 through 22 are not acceptable for use. Nondestructive methods are being analyzed to determine a method in distinguishing acceptable valves from rejectable valves.

NEW VALVE ORDERS

The manufacturing processes have been modified to improve the reliability of new valves:

1. The alloy, C63600, is now prepared by use of a master alloy instead of additions of pure aluminum.
2. The body forgings are inspected by ultrasonic and penetrant methods.
3. The final pressure test of the assembled valve is conducted before the threads of the inlet part are coated with solder.
4. An investigation would be conducted if forgings or valve bodies do not meet acceptance criteria.

The investigations have identified the probable cause of the failures and structural weakness that has made the use of the suspect valves an unacceptable risk. Revisions of the manufacturing processes and the formation of a comprehensive quality assurance plan should insure that new valves are free of material-related problems.

ACKNOWLEDGEMENT

The authors wish to acknowledge the contribution from the staff of the Metals and Ceramics Division, Oak Ridge National Laboratory. The results of the analytical work by electron microprobe and Auger methods was excerpted from *Microstructural Evaluation of UF₆ Cylinder Valve Bodies*, J. M. Vitek, L. Heatherly, and T. J. Henson, Report No. ORNL-6467 (in preparation).

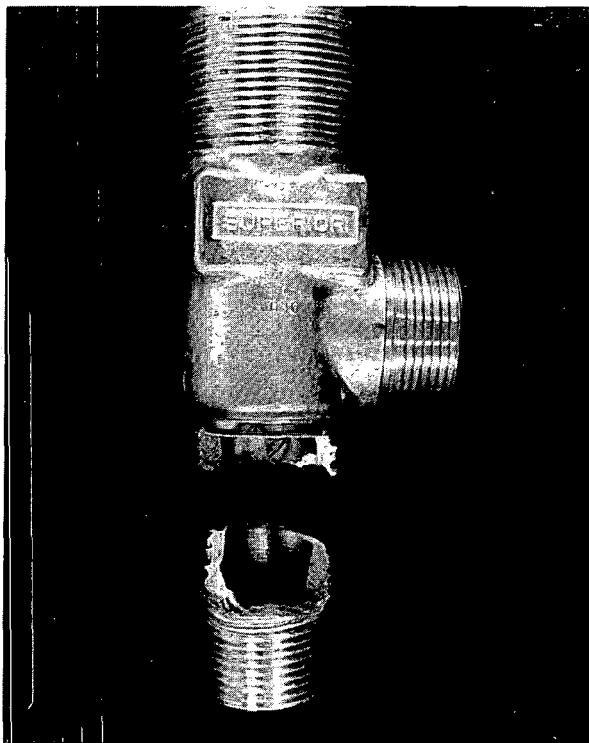


Fig. 1. Valve Body That Cracked After Installation in a Cylinder Coupling and Was Subsequently Pulled to Failure in the Laboratory



Fig. 2. Crack Network Through the Wall of an Inlet Port

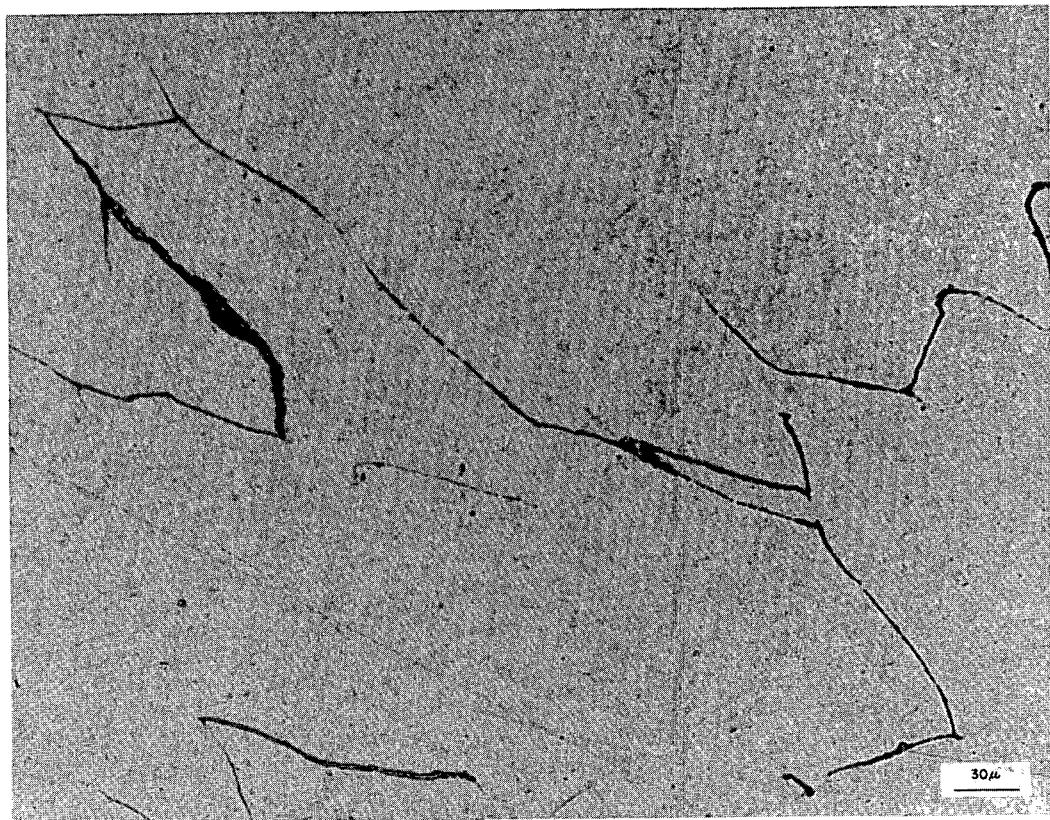


Fig. 3. Network of Grain Boundary Films Near Crack

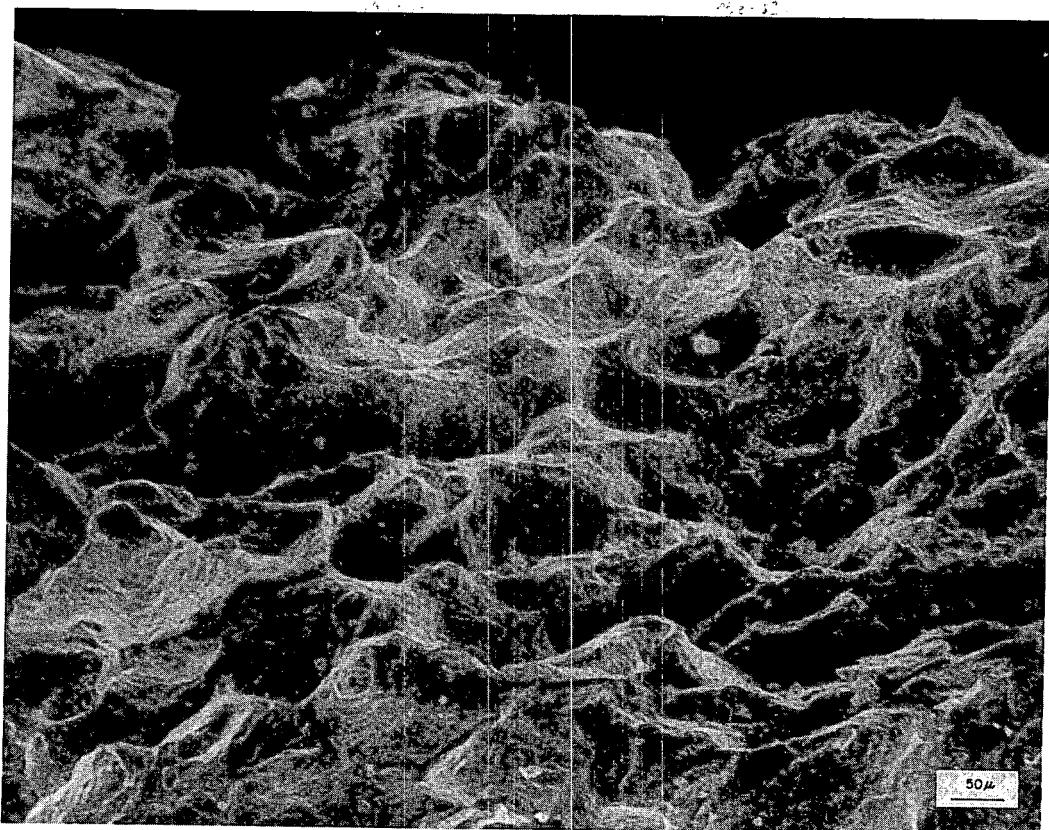


Fig. 4a. Scanning Electron Fractograph of Service Fracture

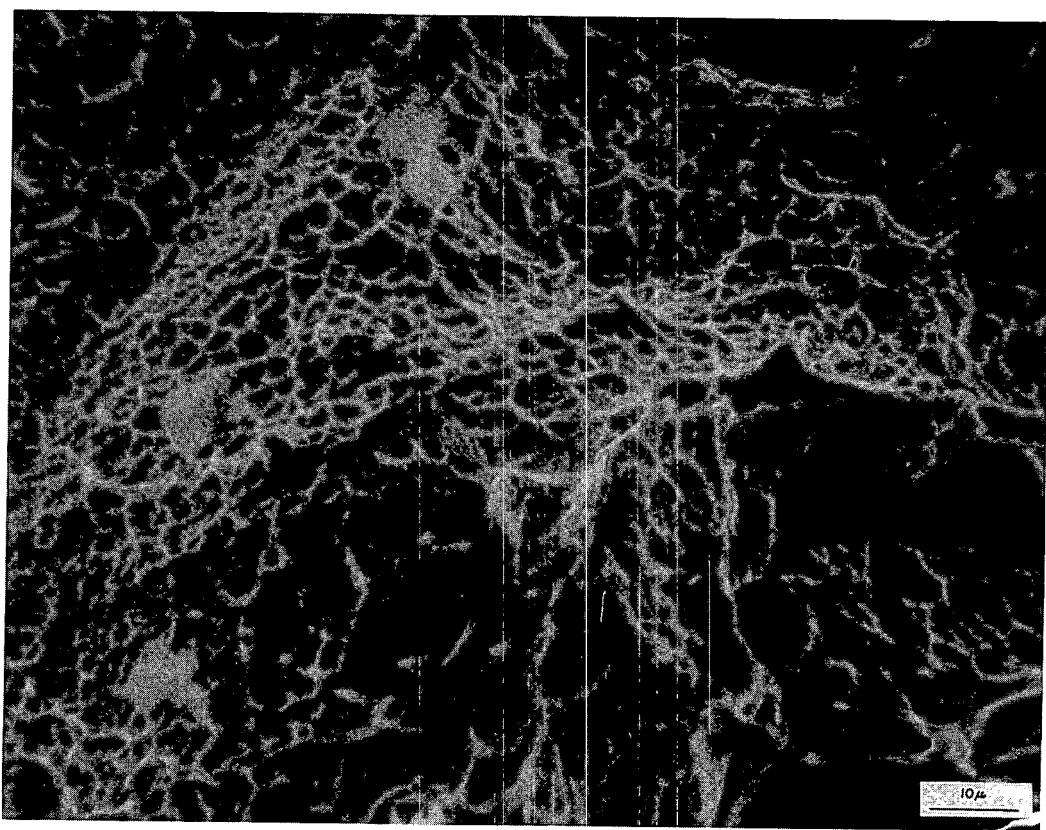


Fig. 4b. Scanning Electron Fractograph of Fresh Laboratory Fracture

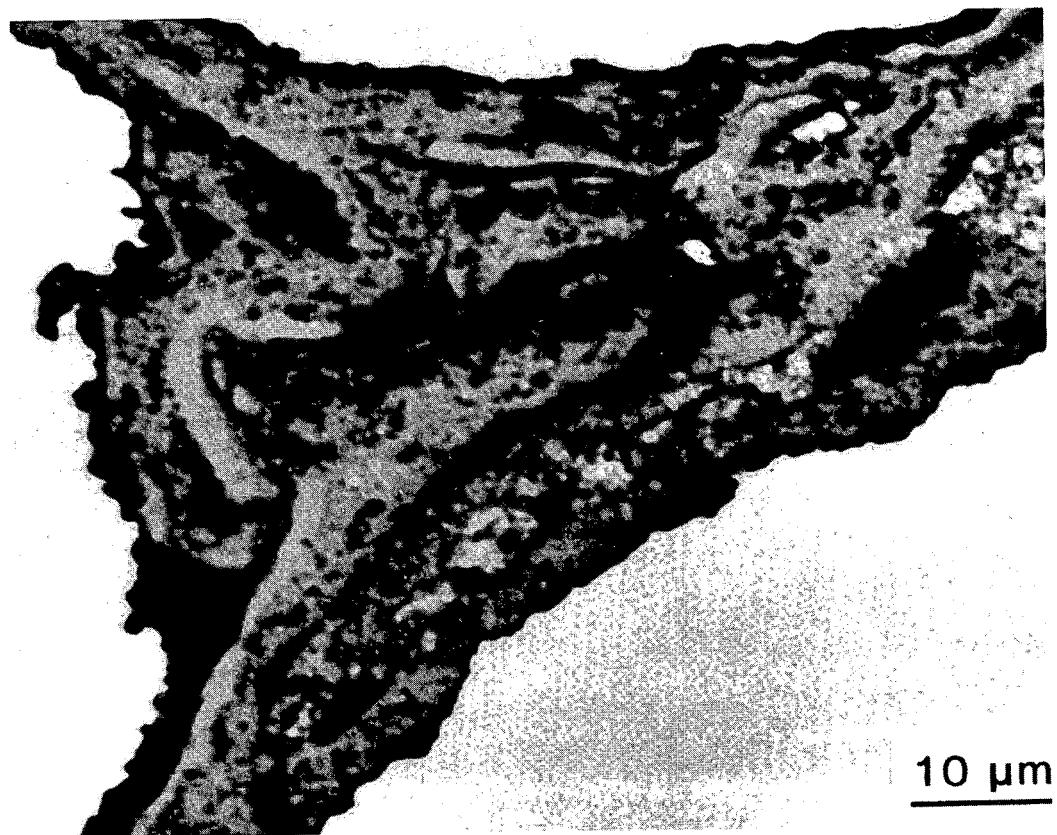


Fig. 5a. Electron Microprobe Backscattered Image of Oxide in Crack

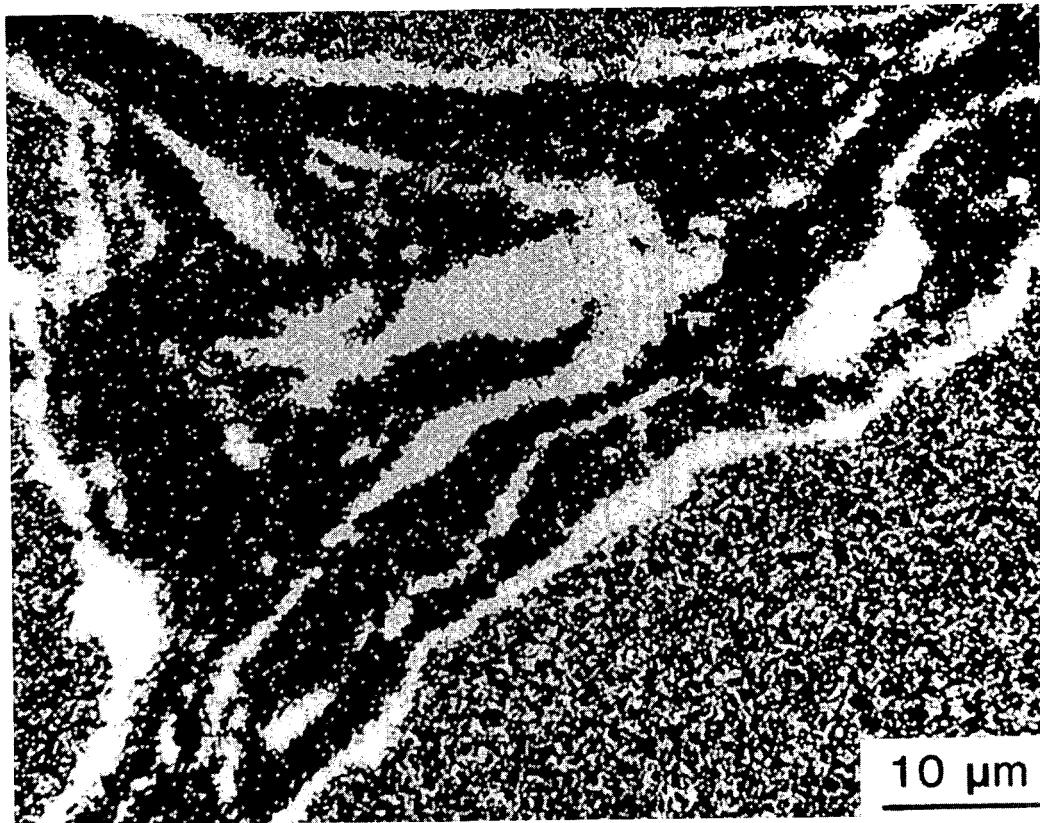


Fig. 5b. Corresponding Elemental Map for Aluminum in Crack

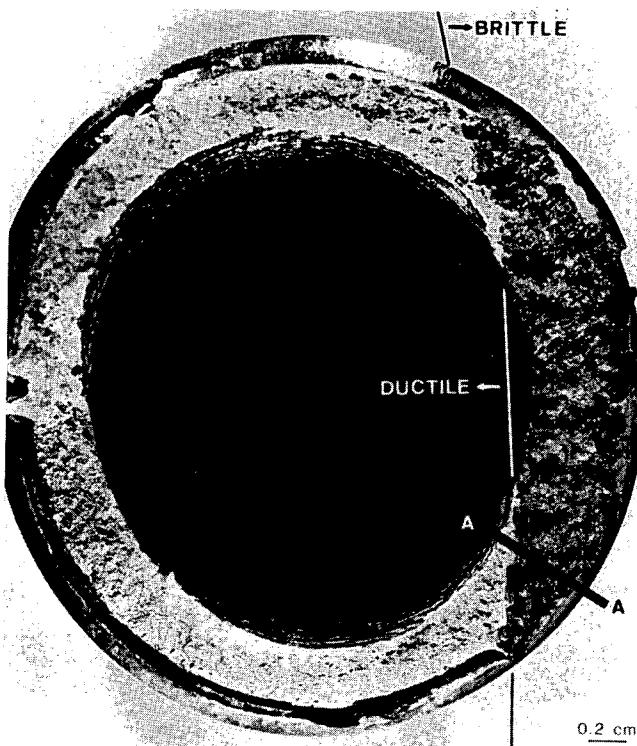
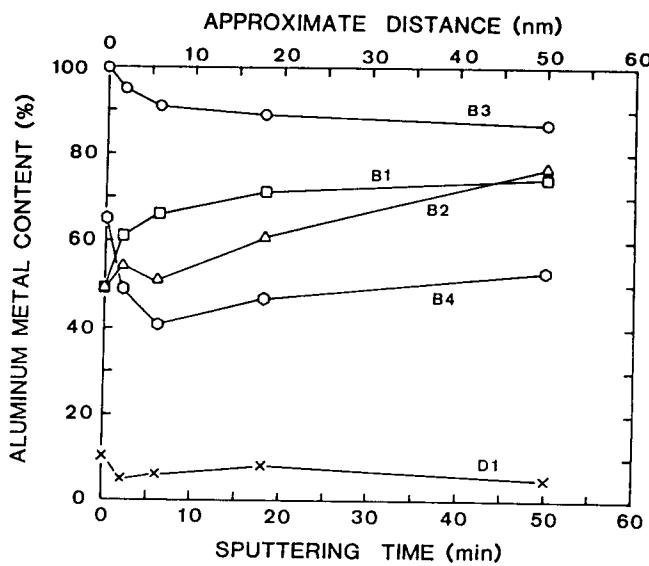


Fig. 6a. Microscopic View of an Entire Fracture Surface Showing the Brittle and Ductile areas

ORNL-DWG 88-6589



The five areas evaluated were:

B1 and B2, dark brittle area; B3, small region within the dark brittle area; B4, lighter, "fresh" brittle area; D1, ductile fracture area

Fig. 7. Plots of Composition Vs. Sputtering Time and Distance From the Fracture Surface of the Aluminum Portion of the Metallic Component

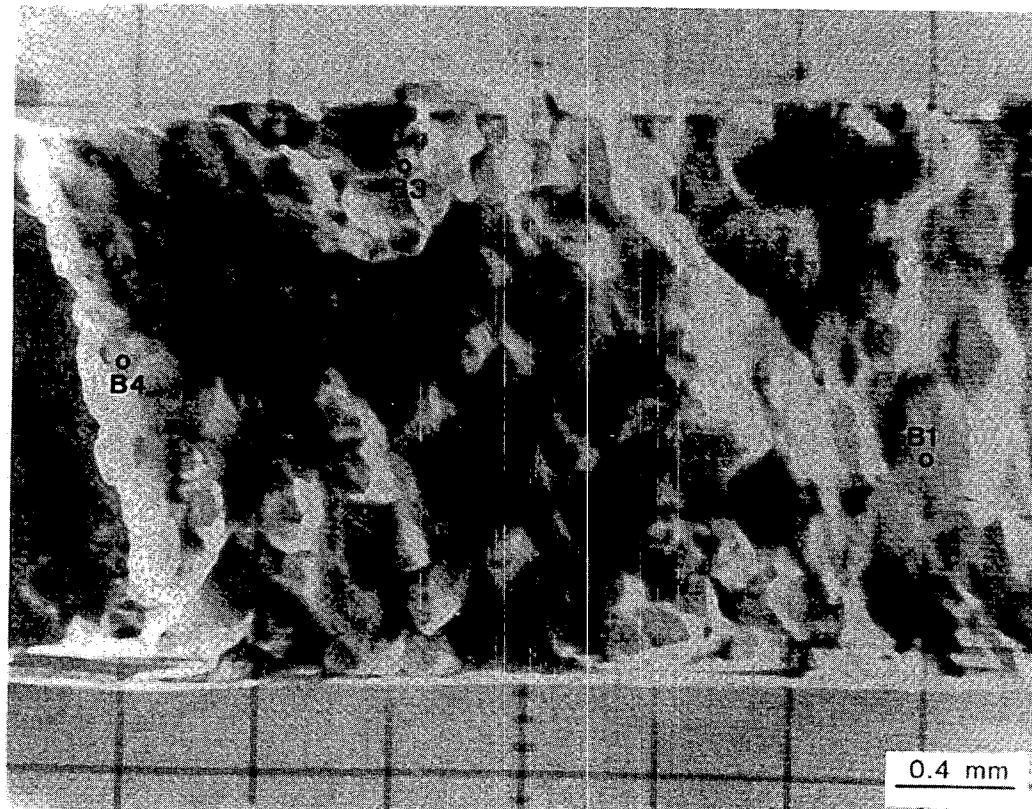


Fig. 6b. Detailed View of the Section A - A of the Fracture Surface in (6a) and Analyzed by Auger Spectroscopy

TESTING OF UF₆ PIGTAILS

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ABSTRACT

The transfer of UF₆ and other hazardous materials between cylinders and manifolds requires the use of pigtails - flexible couplings which allow for misalignment between those parts of the transfer system while providing the required containment of material. To ensure reliability of materials and designs, a UF₆ pigtail test facility was designed and fabricated at the Portsmouth Gaseous Diffusion Plant. This test facility is capable of accommodating the broad ranges of variables found in actual usage. A test is performed by rotating one end of a pressurized pigtail in a circular pattern at the design flexibility requirement until a decrease in operating pressure is detected which denotes failure.

Testing of pigtails has been most beneficial in identifying deficiencies in design, fabrication, and materials of construction. This paper discusses the testing and results of twenty pigtail designs. The resultant uniform fabrication procedures, along with improved inspection methods, have produced safer and more reliable designs.

INTRODUCTION

The flexible coupling used in transferring a liquid or a gas between a manifold system and a cylinder is known as a pigtail. A certain amount of flexibility is necessary to compensate for the normal misalignments between the transfer system and the cylinder. Pigtails for UF₆ transfer at the Portsmouth facility can vary in length from about 18 inches to several feet and are constructed from a variety of materials (tygon tubing, copper, and Monel) depending on service conditions which can range from a vacuum to 100 psi and temperatures up to 250°F. In the late 1970s a corrugated metal hose was proposed to increase flexibility. Since this design change was considered a dramatic deviation from the existing, field-proven, pigtail designs, testing of the design prior to field usage was deemed necessary. A laboratory pigtail test facility was therefore designed and constructed. After the successful initial testing of the corrugated metal hose pigtail design, similar testing of the existing pigtail designs was proposed to document their fitness for intended field service.

The pigtail test facility, Figure 1, was designed to simulate the maximum flexure that the pigtail would experience under normal operating conditions. This is accomplished by offsetting one end of the pigtail to simulate actual usage flexure and then revolving this end of the pigtail through 360°. The rotational speed of the tester is standardized at six revolutions per minute to minimize local heat buildup. During testing the pigtails are pressurized and testing is terminated after a predetermined number of cycles is attained or upon the detection of a decrease in pigtail pressure. The design parameters capable of being investigated are listed in Table 1.

TABLE 1 PIGTAIL TEST FACILITY CAPABILITIES

Parameter	Test Capability
Temperature	Ambient to 300°F (147°C)
Pressure	15 - 600 psia (0.1 - 4.1 MPa)
Angular Offset	0° - 90°
Pigtail Lengths	0.5 - 8 feet (0.2 - 2.4m)
Parallel	
Displacement	4 inches max. (10 cm)
Flexibility	±12 inches max. (±30.5 cm)

A number of safety fixtures have been incorporated into the test system and include safety interlocks installed on the test chamber doors which interrupt the drive-motor power and reduce the system pressure to atmospheric when activated. Other safety features include pressure relief valves to prevent overpressuring, system power interlocks, and an expandable metal, Lexan lined containment chamber to protect against a catastrophic failure.

TESTING RESULTS

Pigtail designs for over twenty different applications have been tested using the pigtail test facility. Several of these designs are shown in Figure 2. The majority of these pigtails were used for the transfer of UF₆, considered as a potentially high risk operation. Three types of deficiencies were discovered as a result of testing these pigtails. The deficiencies were identified as resulting from design, materials, and fabrication techniques.

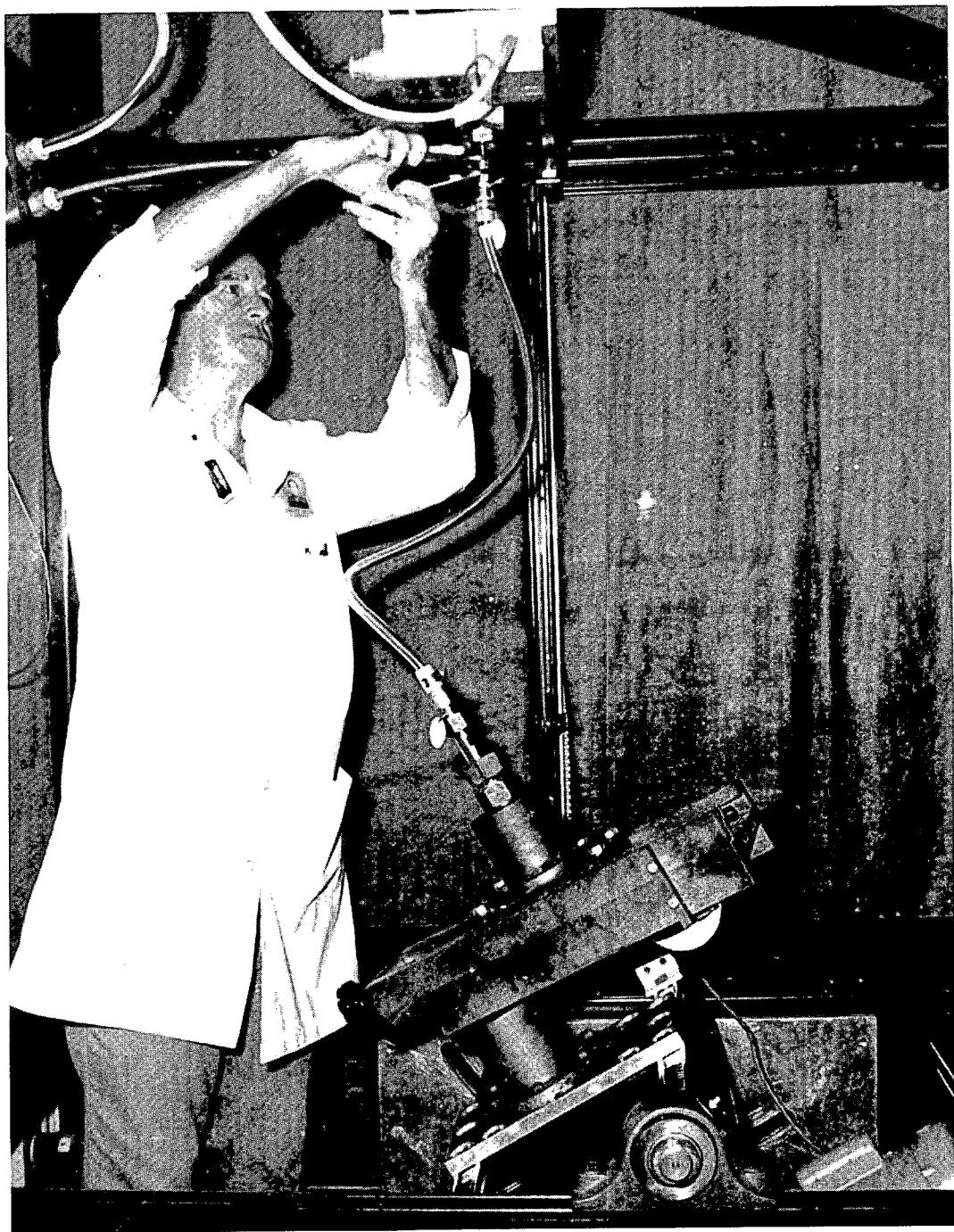
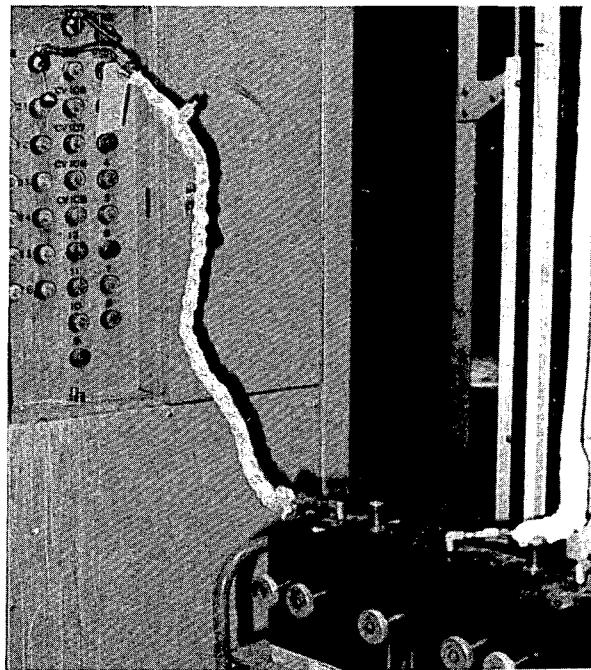
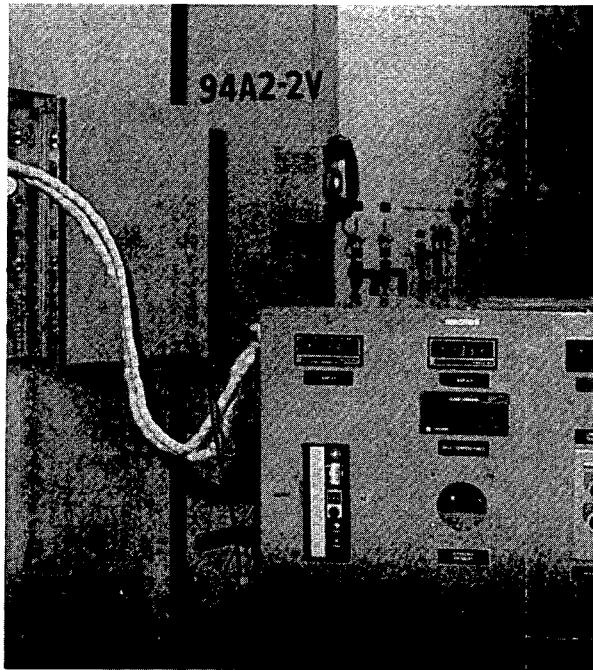
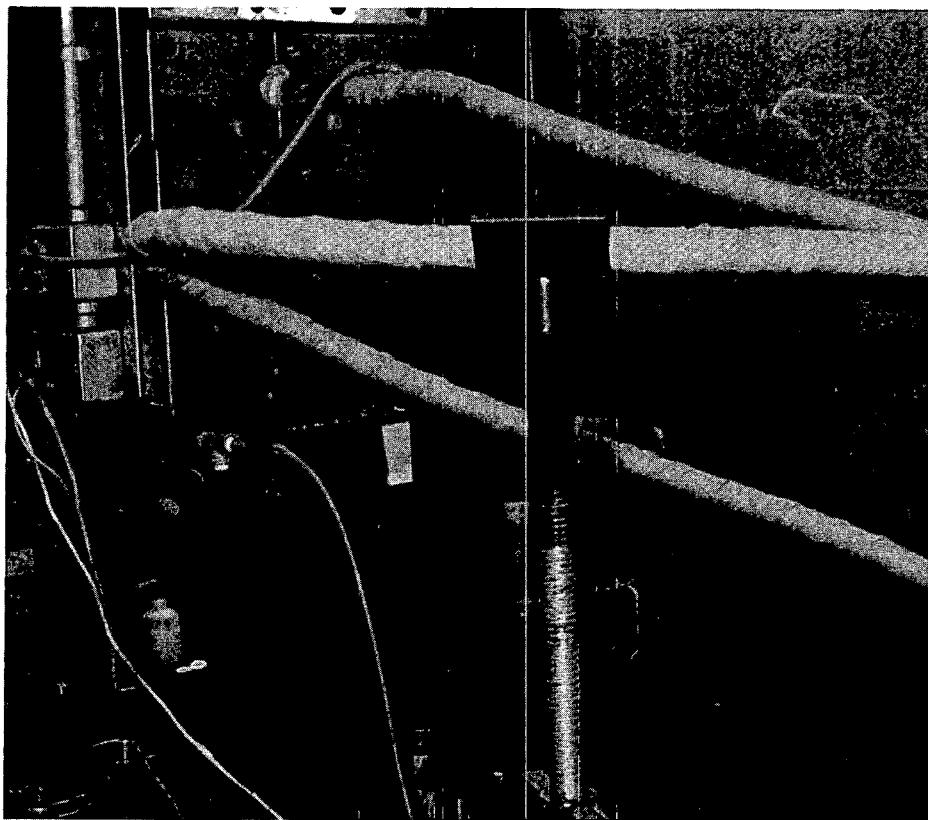


Fig. 1. Pigtail Test Facility Showing Corrugated Metal Hose
Being Installed for Testing



Insulated 3/8" Copper Tubing UF₆ Sampling Pigtails



Insulated 1-1/4" Copper Tubing UF₆ Cylinder Transfer Pigtails

Fig. 2. Examples of UF₆ Pigtails That Have Been Tested

A design deficiency was detected in a prototype pigtail for use in the high assay sampling area (HASA). The pigtail was fabricated from corrugated Monel hose and tested per the parameters for that application. The testing indicated that the pigtail was too short to provide the required flexibility. The pigtail took a permanent set near the rotating end fitting. This "set" could reduce the flexibility needed to obtain satisfactory alignment and sealing during subsequent connections. After adding two inches to the length of the pigtail, the "set" was eliminated, and satisfactory performance was attained.

A material problem was identified during the testing of a copper tubing pigtail. The indenture on the tubing which identified the ASTM Standard of the tubing was too heavily stamped (Figure 3) for the application. The stamping had reduced the wall thickness locally by 14 percent. This reduced wall thickness and the sharp corners of the indentations acted as a notch stress riser and initiated a metal fatigue failure as shown in Figure 4. An inspection procedure and quality assurance actions during tubing procured for pigtail applications have been incorporated to reduce the future risk of receiving unusable material.

The fabrication deficiency that was detected was due to a poorly brazed joint. The braze profile of the tubing-to-fitting connection displayed a concave radius, which resulted in failure at the braze joint during testing. The lifetime of this pigtail was reduced by 75% compared to a similar pigtail design which had a properly brazed connection. Quality Control inspectors were made aware of the potential problem, and the quality of the braze joint has been made part of the inspection criteria.

CONCLUSION

The normal failure mode of pigtails in service is almost always due to abuse, or thread wear due, in part, to cross threading of the fittings. The fittings become cross threaded during hook-up when the misalignment between cylinder and manifold exceeds the design allowance or when the pigtail flexibility is reduced by work hardening or permanent deformation. Normal pressure and vacuum testing as part of the standard operating procedures detect these field failures. Such failures occur at only fractions of the maximum test lifetimes predicted by the flexure testing of adequately designed pigtails. The pigtail test results therefore can not be used to predict field usage life times, but the results have proven successful in identifying design deficiencies before the pigtail was put into field use. The present use of the pigtail test facility is to test pigtails which have had a design change and to aid in determining the cause of premature pigtail failures. The success of this testing system is one additional step in assuring a more reliable transfer of UF_6 and in reducing the risks and vulnerabilities associated with those transfers.



Fig. 3. Variations in Stamping of Copper Tubing for Pigtail Fabrication

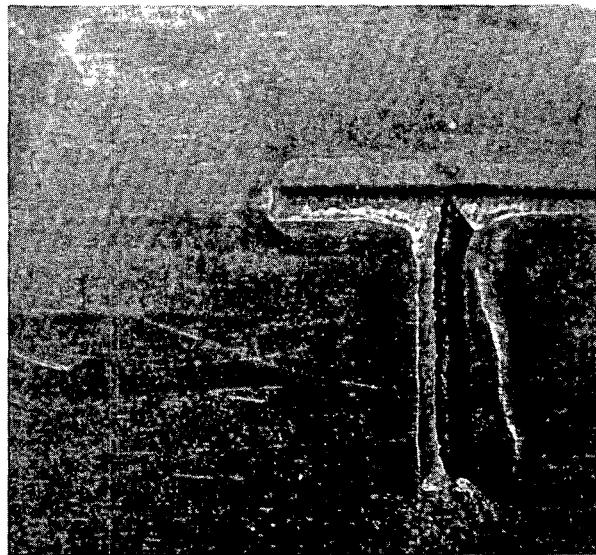


Fig. 4. Crack Initiation in Stamping Indentation

SAFE TRANSPORT OF UF-6 IN THE PRIVATE SECTOR, 1967-1988

BERT JODY, JR.

The topic of my paper, "Safe Transport of UF-6 in the Private Sector, 1967-1988", is a description of the attitudes, technical applications and operational requirements for UF-6 developed by our company, Davis Transport in conjunction with the help and support of the convertors, notably Allied Chemical Company and Eldorado Nuclear, Ltd., over a period of twenty years. This discourse has to do with our own experience with UF-6 and does not purport to cover the entire field of nuclear transportation. It would seem proper to establish the historical background and political climate in which the UF-6 conversion companies came into being, in order to understand those transport decisions which have been made to insure safety.

It is clear that, dating back at least to the Atomic Energy Act of 1954, the general policy of the U. S. Government has always been to encourage and foster the development of a private nuclear industry and to refrain from competing with such industry. The Private Ownership Amendment to the Act is probably the most significant reflection of that long standing policy. That amendment authorized private ownership of "special nuclear material", including uranium enriched in the isotope of U-235. In the 1960's the Act was further amended and criteria published governing the establishment of the Government's massive uranium enrichment program which began on January 1st, 1969. Under this program, customers (mostly domestic and foreign utilities) deliver feed material (UF-6) to the Government's gaseous diffusion plants and subsequently receive therefrom enriched uranium to be further processed into fuel for use in nuclear power plants.

It was in this environment that Davis Transport entered into discussions with Allied Chemical in late 1967 to develop a transport system that would move the first loads of UF-6 under the enrichment program on January 2nd, 1969.

Safety was the subject which overshadowed every aspect of discussions with Allied Chemical which began in the fall of 1967. The transport subjects were basically three in number--the containment cylinder, the trailer structure which would carry it, and the training of personnel who would move the cylinders. First, we discussed the cylinder. A pressure vessel twelve and one half feet long, four feet in diameter, with a loaded weight of 32,000 lbs! This concentrated weight was designed for in-plant applications, but posed real problems in transportation. The cylinder could be deformed, it required a special strong back to lift it, the

cylinder lugs could not be used in the tie down arrangement for fear of damage to the cylinder, and the trailer frame must support the concentrated weight in approximately six feet of length. As if these problems were not enough, the cylinder contents were expensive, corrosive and radioactive if exposed. The cylinders were labeled and placarded radioactive, therefore the public would be involved during transport. Discussions between Davis, Birmingham Manufacturing Co., our trailer constructor, and Allied Chemical established four areas of concern for the trailer. Frame strength center of gravity, suspension and tie down assemblies. Frame strength and its center of gravity were considered concomitantly. The cylinder was to be placed in the horizontal center of gravity over the trailer's main rails at the lowest possible vertical center of gravity to avoid overturn. It was determined that the main frame of the trailer must be able to sustain static weights of two and one half times the weight of the cylinder throughout its length, if we were to have any hope of longevity in operation, the trailer would be a "single drop" platform, with the single loaded cylinder immediately behind the neck transition for load balance and protection from impact. This single drop concept lowered the vertical center of gravity from the nominal 54" deck height of a conventional flat bed to 40 inches of deck height. This one factor reduced the potential for overturn by 14%. The suspension had to reduce the possibility of "road shock" to the cylinder. Spring suspensions on relatively short trailers have a tendency to "crow hop" or lose effective brake time under emergency conditions, and they transmit a good deal of road shock to a fixed load. We, therefore, applied Neway Air bag suspensions to the trailers. Although such suspensions are common today, they were unusual in 1968. It was felt that with automatic leveling valves, the air suspension would lower potential shock to the cylinder and trailer frame and should give better brake application. As it turned out, both suppositions were accurate. These first generation trailers have been in operation for almost twenty years without a single frame failure or weld requirement. In terms of mileage they have each accumulated some 1,700,000 miles without failure.

With the suspension issue resolved, we moved toward solution of the tie down problem. Given the forces which could be exerted on the cylinder in emergency stopping conditions, it was decided that permanent cradles would be welded and bolted into the main frame structure of the trailers. These cradles, two to each cylinder, were of massive 1/2" steel

plate which, when completed, allowed the bottom of the cylinder to sit within three inches of the trailer deck while the ends of the cradle rose some fourteen inches up the side of the cylinder. The load cradles were placed on the trailer in such a way that the outboard stiffening rings of any existing 48 inch cylinder would act to stop forward or aft movement, should it occur. The cradles were twelve (12") inches wide at the mating surface to preclude any "knife edge" effect on the cylinder skin and to provide a large weld and bolt area for securing the cradle.

The upper half of the original tie downs consisted of two 52,000 lb. test wire rope cables which were attached to equal strength clevis and ratchets fastened to the trailer deck thru eye assemblies which were in turn welded into the trailer frame. When assembled and tied down, the cylinders almost became a part of the trailer.

Operational tests were conducted of all the trailer functions at the time of start up. The results were exactly as programmed with the exception of braking and "tracking" of the trailer as a towed unit. In both those cases the results were superior to our expectations. The unit braked with no inclination toward "jack-knifing" and it tracked better than any trailer we had seen at that time. The trailer showed little attempt to "heel over" in tight turns because of the low center of gravity and the automatic leveling valve of the air suspension.

On January 1st, 1969 these units were loaded at Allied Chemical in Metropolis, Illinois and the first cylinders delivered to the Oak Ridge, Tennessee and Piketon, Ohio diffusion facilities on January 2nd, 1969.

The rest, as they say, would be history, except that the nuclear industry was growing. In 1970 we opened discussions for another transport system in conjunction with Eldorado Nuclear, Ltd. of Port Hope, Ontario. Eldorado had a need to ship 48x or 10 ton cylinders, in addition to the 48y or 14 ton cylinders we had been carrying. Was it possible, under existing road weight laws, to carry two 48x cylinders? Working with Birmingham Mfg. Co., we developed a trailer which could do that task without sacrificing the strength factors developed in the first generation trailers. Given the same safety perimeters, we elected to use fabricated main beams of T1 steel. These main rails would have the cradles built as an integral part of the structure inside the rails and the trailer would not have a deck. This trailer somewhat irreversibly was dubbed "slantback" because of its 7% cant from front to rear. The cant, or slanted condition of the trailer was deliberate in that it lowered the over-all vertical center of gravity, especially at the rear. Because of our experience with the previous drop decks, we knew that the tail of the trailer, anchored by a 48x cylinder, would have much less inclination to move away from the center line of travel during a turn. Therefore, the cylinders were placed on the nose of the trailer and the tail of the trailer for load balance and tracking ability. The slanted 7% steel frame permitted static strength in the same range as the original trailers and created a new stopping advantage under emergency conditions. The

cylinders' "G forces" pushed forward but down into the frame, giving the trailer the appearance of "tucking its tail" when the brakes were applied. This factor, coupled with the now standard air suspension, gave exceptional stability to what would otherwise appear to be an unstable structure. This type trailer was put in service during 1970 and 1971 and continues in service to date. No structural failures have occurred in this model during their seventeen year history and 6,000,000 cumulative miles of operation. When one realizes that this unit has an empty weight of only 9200 lbs., it is indeed a remarkable performance.

During the period of time between the second and third generation trailers, the upper tie down units were changed from cable to four inch nylon straps. These nylon restraints were accepted by Allied and Eldorado after some six months of examination of their properties and specific assurances by the manufacturer of their strength. The strapping is certified to 20,000 lbs. per single end strength. This strength is multiplied in that it is attached on both ends. Three (3) tie downs are used to secure the 48y or 14 ton cylinder and two (2) tie downs are used for the 48x or ten ton cylinder. This configuration exceeds DOT restraint requirements. The advantages of this tie down are numerous. First, any damage to the tie down is immediately visible to the driver, loading crew, consignee or regulatory inspector, specifically fraying, cuts or burns. The strapping is impervious to damage by road salt which affected the cable tie downs, and because of its wide, flat, contact with the cylinder, cannot scar or dent the surface. This is especially true when light wall cylinders are to be moved. We have found that certified nylon straps "set" to the load and do not require continuous retightening which occurred with the use of wire rope cable.

In 1980 the U. S. motor carrier industry was favored with deregulation, a recession and fortunately, higher weight limits. During the years of 1980 and 1981, Phillip Smith of Liddel Birmingham Trailer Co. and I collaborated to develop an entirely new vehicle whose purpose was to provide multiple load capability for the nuclear industry. The resultant trailer, called an MVT, incorporated the most positive features of the two previous designs and was constructed under a published Quality Assurance plan. This unit, built of T1 steel was both a drop frame and a "rail" trailer. It has a static weight capacity of better than 3 times loaded weight of 50,000 lbs. and can handle many nuclear packages.

Its primary purpose is to move UF-6 cylinders. The cradles for three cylinders are built into its frame. The main deck height is only 36 inches. The tie down attachments for the certified webbing are close to the cylinders which exerts more holding capacity and more surface contact. The trailer is equipped with disappearing walk boards for handling crews and can be equipped with a winch driver slide which will carry the type B overpack for the enriched 48x cylinder.

The slide can be moved forward or back depending upon the load condition of the overpack. The trailer is equipped with a "plug in" third axle

for loads which exceed standard five axle configurations and can be equipped to move 30b overpacks as well as five ton UF-4 hoppers.

Air suspension of all three axles coupled with low profile tires, allows a lower wider "foot print" for the trailer.

This unit was born out of the never ending quest for safe, versatile, high quality equipment necessary to our industry.

Returning now to the original discussion with Allied and later with Eldorado, the third consideration was training of personnel.

The driver had to be a responsible person who knew how to safely drive a tractor/trailer, attend to all his load related and DOT paperwork, and then what? Was it necessary to educate the driver to the level of a health physicist? The collective decision was in every case "no, we did not need an 'on board' health physicist", but we did need a driver who knew the properties of UF-6, why it was contained in a pressure vessel, and the responsibilities which he, the driver, assumed toward the movement of such material on public highways. Including the driver as a "team member" completed the safety loop in transport.

There is a general assumption that a truck driver, trained to operate a class eight road tractor, can then attach it to any trailer with whatever load it may carry, and deliver the load safely. Nothing is further from the truth. Each type of trailer equipment and load has peculiarities. Each type can be dangerous if not handled properly. A driver must have confidence in his ability to handle the physical dynamics of the trailer which he pulls and confidence in the load strength of the towed unit. When this condition exists, the tractor/trailer becomes an extension of his driving skill and he, the driver, is concentrating on observing the road conditions and traffic around him.

We have always used company drivers and company owned tractors for nuclear transportation. Operational control is better and individual responsibility is promoted through this control.

This paper has, in a very brief way, examined the ingredients of what we have come to call an engineered transport system. The ongoing series of discussions, periods of construction, testing of equipment and the never ending training of personnel have long since welded themselves into a single entity with one purpose, the delivery of UF-6 and other nuclear materials safely, quietly and efficiently. At this point we have moved some 750,000,000 pounds of UF-6 without accident or incident. There are no awards except the business. We promised to do a job and we have done it. From this experience we have made observations and drawn inevitable conclusions for safe operation.

We have observed that all three North American conversion companies have, from inception, used the services of carriers with specialized, dedicated, trailer equipment and trained personnel. Those associations have been long term.

We have observed that those same convertors have

worked closely with their carrier to maintain equipment levels, training and a positive relationship with the public. We have further observed over the past nineteen years of operation that to our knowledge, not one cylinder, loaded on special equipment at the North American conversion plants and destined for delivery to the U. S. diffusion system, has ever been involved in an accident, an accident related overturn, structural collapse, fall from a trailer, or resulted in an injury to a single person during transport. By contrast, we have observed cylinders consigned to the diffusion plants or ports, on systems other than specialized, which have fallen from trains or trailers, caused vans to collapse or overturn, arrive at at least one diffusion plant stacked vertically in a van, and again, jumbled together in gondola cars with their dunnage destroyed. We have observed cylinders with inadequate tie downs and mounted on sub-standard structures which seem to have little relationship to the excellence of the industry which they purport to serve. If this comparison is stark, it is also unfortunately real.

These observations have led our company to certain conclusions.

The basic conclusion is that engineered motor transport systems, incorporating dedicated people and equipment, are the safest means to deliver UF-6 or any other truck load quantity of radioactive material.

An "engineered motor transport system" (E.M.T.) is one in which the equipment has been built to handle the specific loads which it will carry. The E.M.T. system incorporates the personnel of the carrier, the equipment manufacturer, and the shipper in the final decision to submit the end project to the road. Everyone is responsible for its success.

That conclusion is based upon the following facts about an E.M.T. system.

An E.M.T. system is durable. Because it is designed to do a specific, or series of specific jobs, such a system will have the best chance for safe operation over a long period of time. It is therefore dependable. This dependability allows business decisions to be made upon that foundation.

An E.M.T. system is survivable. An engineered system has a better chance of surviving during any emergency condition which might exist.

An E.M.T. system is defensible. It is defensible in law because it is durable, dependable and survivable. The private sector must be ever mindful of the possibility of legal action. E.M.T. systems adopted by the North American convertors, and indeed the U. S. DOD, meet that criteria established in the common law known as "The Act of a Prudent Man". It is prepared to meet that further statement in common law that, "No law or custom, no matter how long in place, may rule to obviate the act of a prudent man".

The E.M.T. system is prudent in that every aspect of operation has been considered before submission to the public highways.

Finally, the E. M. T. system, because it is durable, dependable, survivable and defensible, is also insurable. Insurance is one of the major considerations of any private sector business. Insurance markets for private sector nuclear carriers is narrow and could become non-existent if the insurance industry determines that such carriage is not a good risk.

The E.M.T. system when used as I have defined it, can be tracked by an insurance company to assure itself that risks have been minimized through training, equipment, load restraints and perhaps

most importantly, by attitude. The attitude that safe operation comes before any economic consideration will involve the insurance company as the fourth partner in the E.M.T. system in a positive way.

I wish to thank those persons responsible for this conference and for the opportunity to speak.

I hope that this conference will be considered as a permanent part of the nuclear information calendar in the future.

THE IAEA RECOMMENDATIONS FOR PROVIDING PROTECTION
DURING THE TRANSPORT OF URANIUM HEXAFLUORIDE

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ABSTRACT

The Regulations for the safe transport of radioactive materials⁽¹⁾, are the basis of national and international regulations concerning this subject throughout the world. These regulations require that subsidiary hazards associated with radioactive materials should also be considered. Other national and international regulations concerning the transport of dangerous materials^(2,3,4) consider that a radioactive material having other dangerous properties should be classified as class 7.

Following this line and acting upon the recommendations of SAGSTRAM (Standing Advisory Committee on the Safe Transport of Radioactive Materials) that the Agency should take the lead in providing guidance to Member States with respect to UF₆ packaging and transport, the Agency convened two expert meetings during 1986 and 1987 in order to look into the special problems associated with the transport of uranium hexafluoride. The experts identified several areas in which additional safety measures should be considered if the transport of UF₆ is to have a non-radiological safety level consistent with that of its radiological risks. The recommendations of these groups were published in 1987⁽⁵⁾ and together with comments received were used by another group of experts to prepare a draft of a new safety series document.

In this presentation the new recommendations will be described. The main safety issues to be discussed are fire resistance, valve protection and compatibility with service and structural equipment. Another aspect of importance is the interface between the process and the transport phases, bearing in mind that the same containers are used in both.

This paper will also reveal in how far the new recommendations concerning UF₆ have already been endorsed in the forthcoming European Transport Regulations (ADR/RID) together with the 1985 revised Edition of IAEA Safety Series No. 6.

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5. Recommendations for Providing Protection during the Transport of Uranium Hexafluoride, TECDOC 423 International Atomic Energy Agency, Vienna, Austria, 1987.

**IMPACT IN USA OF PROPOSED
IAEA RECOMMENDATIONS**

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THE REGULATIONS AND THE PROBLEMS OF THEIR IMPLEMENTATION IN UF_6 TRANSPORT

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ABSTRACT

UF_6 is currently transported in packagings which were developed in the sixties — standardized and used all over the world.

In the absence of significant releases during the great number of shipments carried out until now by all modes of transport, even after accidents, we must recognize that these packagings perform their duty adequately.

Nevertheless, we must be aware that the growing amounts of UF_6 issued from URT and the changes in the regulations now raises the problem of compliance of these packagings with the latter.

Among the problems which deserve special attention, the following are particularly noteworthy:

- selection of the packaging type (Industrial, A or B type; fissile or non fissile) in terms of the origin and the enrichment of the UF_6 ,
- design of valve covers (mechanical protection, leakage barrier, criticality control),
- assessment of material behaviour at low temperatures,
- regulatory requirements in handling, tying down cleaning and unloading,
- allowable dose rate increase in case of minor mishaps,
- behaviour in fire, taking into account the toxicity of UF_6 ,
- identification of "special features" required in the case of "controlled moderation" of fissile packages,
- transport conditions of "empty" packagings containing "heels". (LSA or not, allowance of transport without protective packagings).

It seems that most of these points are purely formal and will be easily solved by amendments to the regulations while others are specific to the nature of UF_6 .

It seems also that the desirable modifications to the existing transport packagings are relatively minor, a consensus being necessary on the working conditions.

INTRODUCTION

The operation of the LWR reactors in service in the Western Countries necessitates the annual transport of thousands of 48Y cylinders (each containing approximately 12.5 t of natural or slightly enriched UF_6) and 30B cylinders in their protective packaging (each containing approximately 2.25 t of low enriched UF_6).

Apart from a few accidents and incidents which were minor in terms of their consequences (but which focussed the attention of the public and the authorities on the safety problems involved in the transport of UF_6), the movement of these cylinders did not cause significant pollution until now and therefore the nuclear industry has not felt the need to make any changes to the routines established throughout the world and laid down in ANSI-N14-1-1982 [1].

However, the recommendations concerning the transport of UF_6 (TECDOC-423) [2] published in June 1987 by the IAEA recognized that this standard is not in total conformity with the IAEA-85 regulations [3] or the UNO recommendations for the transport of dangerous goods [4], as result of changes in these two documents (see Table 1).

In view of the unavoidable nature of the transport of UF_6 for supplying nuclear power plants with fuel and the frequencies and tonnages involved, we felt that it was important to methodically identify the differences to which [2] alludes and to propose remedial measures relating to the design of the packagings and the conditions of their use, and even to the regulations themselves if differences are found which do not affect the safety principles on which they are based.

In this paper, we review the results of this analysis, which we have limited to the case of transport using cylinders of 48Y and 30B.

Table 1 – Key dates concerning 48Y and 30B cylinders and the regulations and codes applicable to them

Year	Commissioning	ORO 651	ANSI-N14-1	IAEA S.S.6	UNO ST/SG/AC 10-1
Before 1966	48Y			X	
1966		X		X	
1967	21 PF.1*	X		X	
1968	30B	X			
1970			X		
1971				X	
1972		X			
1973				X	
1974					
1975					
1976					
1977		X			
1978			X		
1979				X	
1980					
1981			X		
1982				X	
1983					
1984				X	
1985				X	
1986					
1987		X	(X)		
1988					

* Publication of Safety Report K-1686

TYPES OF PACKAGINGS REQUIRED UNDER IAEA REGULATIONS AND UNO RECOMMENDATIONS FOR THE USUAL CONTENTS OF 48Y AND 30B CYLINDERS

As shown in Table 2, the 48Y cylinders, being normally used for the transport of natural UF_6 (LSA-1 material), might merely meet the type IP-1 requirements if, as generally accepted, UF_6 may be considered to be a solid. However this requires careful consideration as it is not the case in the transport of emptied cylinders in which more than 80% of the product is in gaseous form at the design temperature ($38^\circ C$). The content is then an LSA-II material and allowance must be made for the type IP-3 if it is wished to avoid "exclusive use" constraints for the return of "empty" packagings. On the other hand, if it is accepted that all movements of 48Y packagings are under "exclusive use" conditions, the type IP-2 requirements suffice even for movements of slightly enriched UF_6 , provided the degree of enrichment is less than 1%, as is at times the case for URT.

It must immediately be stated, as will be explained in detail later on, that the 48Y cylinder in all likelihood belongs to type IP-2 but not obviously to type IP-3, and that consequently the only problem with regard to [3] relates to their transport procedures when "emptied".

Similarly, Table 2 shows that, for 30B cylinders equipped with their protective packagings allowance must at least be made for the type IP-2 requirements (or type IP-3 if freedom from the "exclusive use" constraint is desired), in addition to those for "fissile" packagings.

It must be pointed out that, strictly speaking, they do not need to be of type "A" or of type "B" provided they are systematically checked and cleaned internally, as explained hereafter.

Finally, in view of the corrosive and toxic nature of the UF_6 and the volumes of the 48Y and 30B cylinders, they should also be approved as "tank containers" and meet the requirements laid down for the latter in [4].

Table 2 – Types of packagings required under IAEA regulations for the transport of industrial quantities of UF_6 not more than 5% enriched

Enrichment of UF_6	Cylinders regularly checked and cleaned		Cylinders liable to accumulate residue of low volatility
	Transport "full"	Transport "empty"	
$U5\% \leq 0.72$	IP-1	IP-2 (excl. use) IP-3 (non excl. use)	A
$0.72 < U5\% \leq 1$	IP-2	IP-2 (excl. use) IP-3 (non excl. use)	A
$1 < U5\% \leq 5$	IP-2 fissile	IP-2 fissile (excl. use) IP-3 fissile (non excl. use)	A fissile

PROTECTION OF THE VALVE OF 48Y CYLINDERS

The present valve cover is unnecessarily protruding, and this should be avoided for conformity with [3]. In addition, not only is their correct installation difficult to check but we now know the risk of damaging the valve during makeshift handling operations for remote recovery of cylinders (wreck of the Mont Louis).

Finally, tests showed that they do not guarantee leaktightness of the valve in the case of a full cylinder falling 0.6 m onto a rigid surface as provided in §§5.3.7 and 6.2.2 of [3].

A large number of solutions to these problems have been proposed and all have their advantages and disadvantages. Certain are extremely simple and inexpensive. But if it is desired for the 48Y cylinder to be considered to be of the IP-3 type and thus usable for "emptied" return transport and not necessarily in "exclusive use", it is necessary for the cover to also constitute an "enclosure capable

of retaining any leakage from the valve", which is not currently the case.

For instance, the cover and its attachment to the cylinder could be of the type schematically shown in Figure 1.

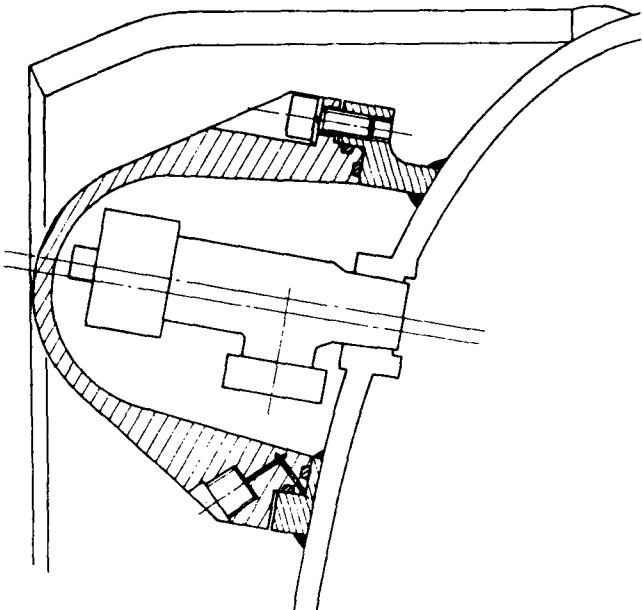


Figure 1 – Example of installation of a leaktight valve cover on a 30B cylinder

In this example, the cover is provided with a flange equipped with a gasket (preferably double) tightened by screws to a backflange welded to the body of the cylinder.

Such a modification of the existing cylinders could be carried out on the occasion of their periodic inspection by the authorized inspector as it is compatible, without post-weld heat treatment (PWHT), with the requirements of ASME-Section VIII [5].

This would compensate for the unsophisticated nature of the valve (which is much appreciated), or incorrect use of it, by constituting a reliable and easily verifiable barrier capable of containing any leakage at the valve and providing the valve with effective protection against impact.

PROTECTION OF 30B CYLINDER VALVES

A distinction must be drawn between the case of "full" and "emptied" cylinders: "emptied" cylinders, which can contain as much as 11.35 kg of 5% enriched UF_6 , can be transported without protective packaging, as provided under §§8.1 and 8.2 of [1].

Nevertheless, it is clear that the valve cover is not designed to contain any leaks from the valve and, in addition, that its ability to protect the valve in the event of a 1.2 m drop onto a hard surface or

the impact of a 6 kg bar falling from a height of 1 m remains to be demonstrated in the case of a bare 30B cylinder.

It is for this reason that a cover of the type already proposed for the 48Y cylinders would be desirable to enable the cylinder to meet the regulatory requirements applicable to IP-2 and IP-3 packagings.

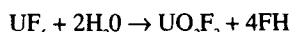
The 30B cylinders containing more than 11.35 kg of UF_6 must be transported inside a protective packaging such as the 21 PF.1 type. This assembly constitutes a "fissile" package of which the nuclear safety and the approval are based on the "controlled moderation" principle.

Indeed, the authorized content of the package is limited to 2,279 kg of 5% enriched UF_6 , whereas the minimum volume of a 30B cylinder is 736 litres.

As the density of UF_6 is close to 5.1 g/cc at 20°C, within a 30B cylinder containing the maximum authorized load there will remain a void (plenum) of which the volume at 20°C will be at least:

$$736 - \frac{2279}{5.1} = 289 \text{ litres}$$

Should water enter this plenum, it will react with the UF_6 in the following manner:



At 20°C, UO_2F_2 and FH are respectively a solid and a liquid, both being soluble in water.

Consequently, if a 30B cylinder with a leak is immersed in water at a depth of 15 m as provided in [3], its internal moderation is only limited by the quantity of hydrogen in the water liable to occupy the volume of the plenum, i.e.

$$289 \times \frac{2}{18} = 32 \text{ kg}$$

This is considerably more than it is necessary to attain the critical state, which for a homogeneous solution of a salt of 5% enriched uranium corresponds to masses of uranium and hydrogen respectively close to 45 kg and 4.8 kg.

For such a "fissile" package of which the nuclear safety is not guaranteed in the event of ingress of water (or loss of the content), §5.6.5 of [3] also imposes, in addition to general requirements applicable in the design and utilization of all "fissile" packages, "special features" identified and recognized by the competent authority responsible for approving the concept as preventing any leakage, even resulting from human error. [3] also requires that these "special features" be specified in the approval certificate, clearly to draw the attention of the other competent authorities to this particularity, subject to validation.

As [3] notably considers that "multiple high standard water barriers" together with reliable leak tests permit compliance with this "special features" requirement, the present packaging (30B cylinder + 21 PF.1 protective packaging) could be equipped for instance with a valve cover of the type shown in Figure 1.

FIRE RESISTANCE

[2] raises the question of the fire resistance of packages containing UF_6 as requested by UNO recommendations [4] for "tank containers".

We consider that this question does not involve any major difficulties as it is likely that the designs of the 48Y and 30B cylinders are acceptable in view of the conditions (30 minute fire at 650°C) and the criteria (allowable leakage at valve) specific to these recommendations.

Concerning this subject, we note that:

- "Emptied" 48Y and 30B cylinders are not liable to explode in view of the limitation of their authorized content which corresponds to extremely low pressures at 650°C and which, even if no allowance is made for leakage at the valve, would not exceed 1.2 bar for the 48Y cylinder and 3.3 bar for the 30B cylinder
- Neither are "full" 48Y cylinders liable to explode due to their thermal inertia and to the volume of their plenum, as made clear in other papers presented at this conference.

The problem is limited to the case of partly filled cylinders for which there is a risk of rupture under the effect of the pressure attained by gaseous UF_6 . There is a need to specify the allowable filling ranges, which should be indicated in the utilization procedures and other reference documents such as [1] and [2]. Provided such limits are met, the 48Y cylinder itself would not require modification to guarantee its fire resistance.

HANDLING, STABILIZING AND TIE-DOWN FITTINGS

§5.0.5 of [3] requires that the design of the packagings makes it safe and easy to handle them and tie them down. In addition, [4] requires stabilizing members designed, together with the tie-down fittings, to resist an acceleration of an intensity of $[(2)^2 + (2)^2 + (1)^2]^{1/2} = 3G$ (without exceeding 2/3 of the yield strength).

We observe that these stabilizing members are absent from the 48Y cylinders which do not even always have means allowing their handling when fastened to a transport chassis.

To remedy these shortcomings, it would be sufficient to ensure universal use of approved transport frames and to provide the cylinder handling lugs with additional eyes for tying them down to these frames.

The unit could then fully comply with the requirements set forth in [4] for "tank containers" (see Figure 2).

The 30B cylinder does not have a base either when it is shipped bare and, as in addition it does not have tie-down fittings and as its handling method is not obvious, the question is raised as to whether the use of protective packaging should not be extended to empty 30B cylinders as well. Furthermore, the conformity of the handling and tie-down fittings of the protective packagings with regard to [4] should be checked.

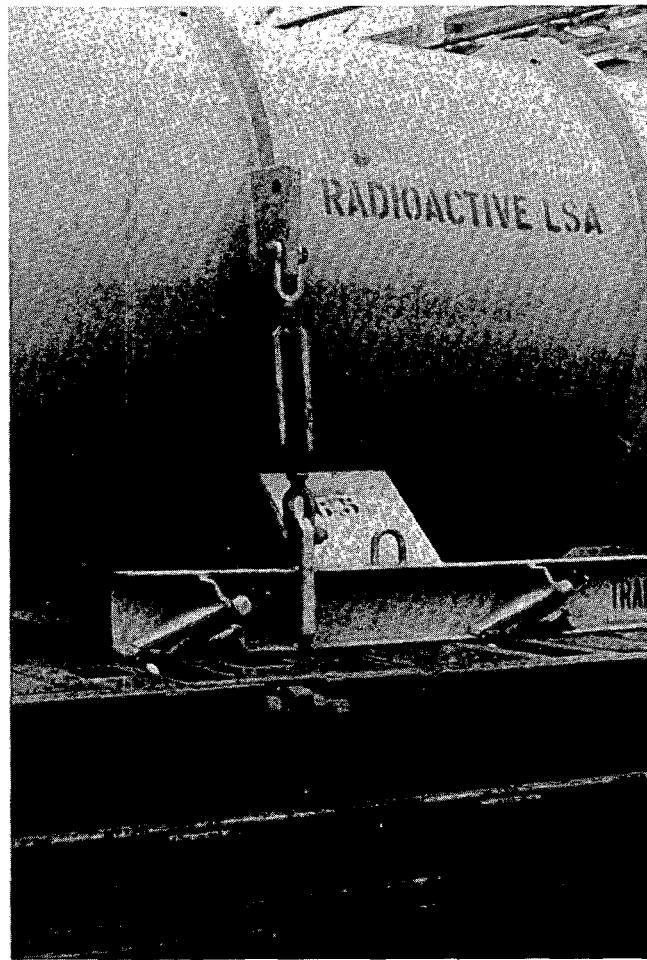


Figure 2 - Handling lug modified to allow handling of 48Y cylinders fastened to their transport frame

PROBLEMS RELATING TO THE ACTIVITY CONTAINED IN THE CYLINDERS

Up to 5% enrichment, UF_6 is considered to be an LSA material as the activity limit A_2 attributed to the uranium up to this degree of enrichment is infinite and the definition of "non irradiated uranium" given by [3] makes it possible to ignore any radioactive impurities contained in commercial UF_6 below limits which actually go beyond those specified by the industry for UF_6 issued from URT as well as UNAT.

However, as cylinders are often not completely emptied after unloading, it is possible that there may be a build-up inside them of compounds of low volatility of which the activity depends on the origin and degree of purification of the UF_6 , as well as the time for which it has remained in the cylinders since their last cleaning. As a matter of fact doses in excess of 2 mSv (200 mR/h) have been recorded in contact with 48Y cylinders "emptied" of their URT content. In this case, the residual content of the containers is obviously not a chemical compound of "non irradiated uranium", and, as the material is no longer LSA, its transport would necessitate at least a "type A" packaging. It is thus clear that to determine which type of package should be adopted, it would be necessary to know the maximum quantities of impurities liable to be found in

"emptied" cylinders. In reality, if we assume that the cylinders already satisfy the requirement concerning IP-3 packagings, they may be considered to be "type A" if the materials of which they are made perform their functions at -40°C, the only requirements which practically differentiates these two types of packagings. As concerns the 48Y and 30B cylinders, we can consider that this requirement is met as they are made of ferritic steel sheet of which the thickness ($\leq 5/8$ of an inch), the grade (SA516 grade 55, 60 or 65), the steel making practice (fine grain practice) and the heat treatment (normalized) guarantee an ample margin of safety with regard to the risk of brittle fracture, if reference is made to §5.2.3 of recommendations NUREG/CR 1815 [6].

As mentioned earlier, the low volatile impurities in the UF₆ accumulating on the inside surfaces of "emptied" cylinders create a radiological problem which can only be solved by imposing frequent dose rate checks and thorough cleaning, as a function of criteria which remain to be determined. The last consequence of these radioactive deposits results from the requirement in [3] of not risking an increase of more than 20% of the dose rate in contact with "type A" (or IP-3) packages after tests representative of normal transport conditions (free fall of limited height, compression and bar penetration). If we accept the presence of radioactive deposits on the inner wall of 30B cylinders, it would be necessary to transport them in a far more rigid protective packaging than the type 21 PF.1 if this requirement is to be met. Cleaning of the cylinders cannot suffice to solve this somewhat formal problem, which in our opinion calls for revision of [3].

RECOMMENDATIONS

In view of the large number of cylinders in existence, it would be desirable for the 48Y and 30B packagings, on which the safety of industrial transport of UF₆ depends, to remain operational for a long time to come.

Nevertheless, this requires changes in the design of these packagings and the conditions of their use, which were established before the current regulations evolved.

Provided our interpretation of requirements of [4] concerning fire resistance is correct, these changes should presumably be minimal as concerns the packagings themselves. The principal problem relates, in our opinion, to the valve protection covers, which should have greater resistance to impact and constitute a verifiable leakage barriers.

As concerns the operational procedures which require modification, we consider the most important to be more stringent monitoring of leaktightness (particularly for 30B cylinders), filling limits for "full" 48Y cylinders, the compulsory use of a transport frame of an approved model for these cylinders and the flushing of the interior of cylinders which during transport have or are liable to have a dose rate in contact or at a distance which exceeds the regulatory limits.

Finally, further to this exercise in analysis concerning the conformity of the 48Y and 30B cylinders with the regulations, it would be desirable to recommend to the IAEA to reexamine their recommendations as specifically concerns:

- certain definitions (non-compressed gas and non-irradiated uranium),
- the conditions to be fulfilled to satisfy the controlled moderation principle,
- the requirements concerning handling and tying down (is the absence of specific attachments acceptable?),
- the 20% increase of the dose rate in contact with packages after "type A" tests.

In view of the importance and urgency of these modifications to the regulations and of their possible effect on the transport of other radioactive materials, it would no doubt be better to apply specific packaging rules to UF₆. These should, insofar as possible, be intended to perpetuate the use of the 48Y and 30B packagings without making concessions with regard to safety principles (risks of release, controlled moderation, handling and tying down) as generally applicable to the transport of dangerous goods and, more specifically, radioactive and corrosive materials.

REFERENCES

- [1] American National Standard for Packaging of Uranium hexafluoride for transport, ANSI N14.1, 1982.
- [2] Recommendations for Providing Protection during the transport of Uranium Hexafluoride, IAEA, TECDOC-423 (1987).
- [3] Regulations for the safe transport of Radioactive Material, 1985 edition revised in 1986, IAEA, SS No. 6 (1985) and IAEA SS No. 6, Suppl. 1986 (1987).
- [4] Transport of dangerous goods. Recommendations of the Committee of Experts on the Transport of Dangerous Goods, ST/SG/AC.10/1 Fifth revised edition (1988).
- [5] ASME Boiler and Pressure vessel code, Section VIII, Div. 1, 1986 edition.
- [6] Recommendations for Protecting Against Failure by Brittle Fracture in Ferritic Steel Shipping Containers Up to Four Inches Thick, NUREG/CR-1815 (1981).

THE CLEANING OF URANIUM HEXAFLUORIDE CYLINDERS CONTAINING
RESIDUAL QUANTITIES OF URANIUM HEXAFLUORIDE AND IMPURITIES

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ABSTRACT

There is a need to clean cylinders which have contained natural uranium in the form of uranium hexafluoride at the Metropolis, Illinois plant of Allied-Signal, Inc. The intent of the cleaning procedure is to neutralize and remove residual UF₆ and nonvolatile impurities and daughter products which accumulate over a period of time. It should be emphasized that this procedure is applicable to cylinders which have been used to contain only natural uranium and in which the residual material totals less than fifty pounds.

The cleaning procedure is begun by connecting the cylinder to a vacuum source which is supplied by circulating a sodium carbonate wash solution through an eductor. After the UF₆ fumes have been removed from the cylinder, the interior wall of the cylinder is sprayed with the wash solution which is maintained at an appropriate pH to solubilize the uranium. The pressure in the cylinder is carefully regulated during the chemical reaction phase of the cleaning procedure. After the washing has been completed, the contents are drained from the cylinder, and the cylinder is rinsed with water. The sodium carbonate solution is filtered and then processed for the recovery of uranium. The insoluble material containing the daughter products is removed by the filtration.

TEXT

The primary business of Allied-Signal, Inc. at its facility located at Metropolis, Illinois is the conversion of uranium ore concentrates into uranium hexafluoride. The UF₆ cylinder handling requirements are considerable at this location. While some cylinders filled at the Metropolis Plant are customer cylinders, Allied fills, ships, and maintains mostly its own cylinders. Hydrostatic pressure testing of these cylinders is performed at regular intervals as required by regulations. Hydrostatic testing necessitates the cleaning of UF₆ cylinders, prior to the performance of the test. On occasion "crude" UF₆ or out of specification material may be filled

into a cylinder, and sometimes cylinder maintenance is required. Either of these situations may also require that cylinders be cleaned. The cleaning is accomplished by spraying the interior of the cylinder with a sodium carbonate wash solution which is maintained at an appropriate pH. The cylinder is then rinsed, steamed, and dried to complete the cleaning process. Insoluble impurities are removed by filtration. All personnel who use this procedure have been locally trained and qualified.

The cylinder cleaning procedure serves to neutralize and remove residual UF₆ and nonvolatile impurities and daughter products which accumulate over a period of time. The primary nonvolatile impurity is in the form of molybdenum fluorides. The daughter product of primary concern is the fluoride of the Thorium-234 isotope. This paper does not address chemical or radiological safety except to state that those are concerns which the user of such a procedure must consider. He must then administer necessary safety precautions. After the cylinder has been cleaned, it is available for visual internal inspection and subsequent testing and/or maintenance. The basic procedure is described in the following section. A drawing (Figure 1) has been included to clarify the steps of the procedure, and it is referenced throughout the description which follows.

PROCEDURE

1. Place the cylinder in the cylinder cradle with the valve in the 12 o'clock position. The cylinder is positioned so that the cylinder plug is toward the end of the cradle nearest the sump.

NOTE: Throughout this procedure it is necessary that appropriate safety equipment be used and safety attire be worn.

2. Secure the cylinder to the cylinder cradle.
3. Batches of sodium carbonate solution are

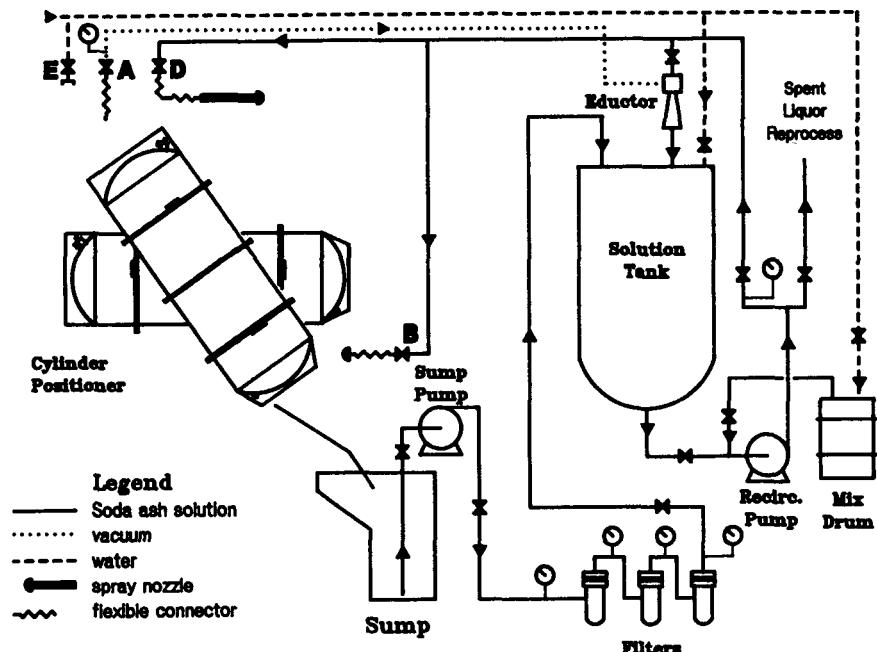


Figure 1.
Cylinder Wash Flow Diagram

prepared in a mix tank and pumped to the solution tank to provide a wash solution of pH greater than 9.0.

4. After the solution tank has been filled with the wash solution, open the tank drain valve and the valve inlet the eductor on top of the tank, and start the recirculating pump. Valves A, B, and D are closed prior to starting the pump. This establishes flow through the eductor and provides a source of vacuum.
5. Check the pressure gauge at valve A. The gauge should read 26 to 30 inches of mercury vacuum. With the cylinder valve closed, connect the flexible connector from valve A to the cylinder valve. Open valve A, and then slowly open the cylinder valve.
6. Evacuate the cylinder to 26-30 inches of mercury vacuum and hold at that pressure for at least fifteen minutes. This removes fumes from the cylinder and neutralizes them as they contact the basic solution passing through the eductor.
7. After the prescribed time period has expired, remove the cylinder drain plug, and attach the spray wash adapter to make a pressure tight connection.
8. Attach the hose from valve B to the adapter and open valve B.

9. Valve B should be regulated to maintain a pressure on the system of about 10 inches of mercury vacuum. If the pressure, which occurs as the result of chemical reactions within the cylinder, increases faster than it can be evacuated, close valve B. The eductor and pump system should be used to deliver about 120 gallons of solution into the cylinder.
10. Close valve B and raise the plug end of the cylinder so that it is slightly higher than the valve end of the cylinder.
11. Remove the spray wash adapter and replace it with a drain valve.
12. Rock the cylinder back and forth to distribute the solution throughout the cylinder. This is the primary washing action.
13. Close valve A and disconnect the evacuation line. Remove the cylinder valve. The cylinder valve should be higher than the drain valve during the performance of this step.
14. Raise the cylinder valve end of the cylinder to the near vertical position. Attach the sump hose to the drain valve, open the valve, and drain the contents of the cylinder to the sump.

15. Start the sump pump. Check the pH of the wash solution in the solution tank and maintain it above pH = 8.0, adding additional sodium carbonate if necessary.
16. Check the pressure drop across the bag filters. When they become blocked, they must be replaced. (Filter bag replacement and disposal are addressed in a separate procedure).
17. Place the spray nozzle in the cylinder valve opening. Attach the rubber hose from valve D to the spray nozzle, and open valve D. This is the secondary washing action.
18. Close valve D, and rinse the cylinder with water supplied at valve E. Continue the rinse until the solution draining from the cylinder is clean.
19. Close valve E, and steam through the cylinder until the walls are hot. Dry the cylinder immediately by purging dry air through the cylinder.
20. Visually inspect the inside of the cylinder from both ends. If the cylinder is not clean, the procedure must be repeated.
21. Shut down the recirculating pump. When the sump is empty, shut down the sump pump.
22. The material in the solution tank is pumped to a uranium recovery process as required.

Future activities involving the cylinder will depend upon whether it is to be hydrostatically tested, repaired, or returned to service.

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CLEANING AND INSPECTION EXPERIENCE OF UF₆ 30B CYLINDERS

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ABSTRACT

Operational experience on cleaning and inspection of UF₆ 30B cylinders started in 1985 is described.

Cleaning results show that surface radiation rate of cylinders ranging from 1 μ Sv (0.1 mrem) ~ 0.7 mSv (70 mrem) per hour before washing is reduced to 0.2 μ Sv (0.02 mrem) ~ 1.5 μ Sv (0.15 mrem) per hour after washing. Average weight of wet precipitate containing uranium is less than 3 kg per cylinder, which are higher than originally expected. This precipitate is put into polyethylene packs, which contain some 3 kg per pack in average and whose surface radiation dose rate ranges from 0.07 mSv (7 mrem) to 0.85 mSv (85 mrem) per hour. The average amount of process liquid waste is approximately 0.6 m³ per cylinder, and after treatment its α and β radioactivity become $<7.4 \times 10^{-3}$ Bq (2×10^{-7} μ Ci)/cm³ (α) and $<1.1 \times 10^{-1}$ Bq (3×10^{-6} μ Ci)/cm³ (β) respectively.

Radiation control results such as personnel dose rate, airborne concentration in the restricted area and that of gaseous effluent from the stack and so on are also presented. The radiation dose rate of the operators is very low in both whole body dose and skin dose.

1. INTRODUCTION

When UF₆ cylinders are evacuated, their surface radiation dose rate due to residual uranium daughter becomes so high that they can not be transported for refilling UF₆ immediately after evacuation because of Japanese Regulation for transportation.

In order to solve this problem and to minimize cylinder inventory, it was decided to have a cleaning and inspection facility of 30B cylinders. Then, based upon preliminary washing tests and various studies and investigations performed for several years, the facility was designed and constructed in 1983, the system concept of which depended basically upon the procedures stated in ANSI N14.1 and ORO-651. Total process consists of receipt and check of cylinders to be cleaned, cleaning, inspection, and processing of wash solution including precipitate and residue handling and liquid waste treatment.

To establish operating conditions, 10 empty cylinders were washed for washing and inspection tests. As a result of these tests, cleaning operation could be started in June, 1985 and since then approximately 340 cylinders have been washed to date. And in April, 1987, periodic inspection due every five years could be started in our plant and the periodic inspection of some 70 cylinders have already been completed.

2. PROCESS OUTLINE

As the maximum surface radiation dose rate of the cylinder (normally at the bottom) soon after vaporization in conversion area is very high, it is left in the restricted area until radiation dose rate decreases low enough to avoid excessive personal exposure. In the cleaning facility the criteria for surface radiation dose rate are 1 mSv (100 mrem)/hr, that is, only the cylinder below 1 mSv (100 mrem)/hr can be washed.

In Fig. 1, Total Process is shown, that is, from receipt of cylinders to shipping of the washed and inspected cylinders.

Receipt and Check Flow Chart is shown in Fig. 2, in which check items for the cylinder to be washed are described. The most important items are the radiation dose rate and the cylinder weight, which are compared to those of the cylinder after washing. Actual heel can be obtained as the difference between the weight of the cylinder before washing and that after washing.

Fig. 3 shows Cleaning Flow Chart and in Fig. 4 Shower Washing seen in Fig. 3 is schematically described. Uranium concentration of the 2nd wash solution is measured and in case it exceeds 500 ppmU, both shower washing and 2nd water washing are applied again. When installing a valve, a valve thread engagement of 7 minimum and 12 maximum is applied by using a minimum of 2770 kg·cm (200 foot-pounds) and a maximum of 5540 kg·cm (400 foot-pounds) of wrench torque as described in ANSI N14.1.

In Fig. 5 Treatment of Wash Solution consisting of precipitate and residue handling and liquid waste treatment is shown.

Fig. 6 shows Inspection Flow Chart, which includes Routine Inspection and Periodic Inspection. Periodic Inspection is observed and approved by a Qualified Inspector specified in ANSI N14.1. To date none of the cylinders inspected, which amount to some 70, have been rejected.

3. RESULTS OF OPERATION

3.1 Cleaning results

More than 340 cylinders have been washed since the cleaning operation was started. In Table 1 actual washing data are listed and as shown in the Table design capacity of one cylinder per day has been achieved.

Average heel weight is approximately 380 g per cylinder, though range of the weights is rather wide.

Surface radiation rate of cylinders before washing ranging from 1 μSv (0.1 mrem)/hr to 0.7 mSv (70 mrem)/hr becomes 0.2 μSv (0.02 mrem)/hr ~ 1.5 μSv (0.15 mrem)/hr after washing.

Average weight of precipitate is about 2.6 kg per cylinder, which are much higher than expected value originally designed. It is necessary for us to make an effort to reduce this value. This precipitate is put into polyethylene packs, which contain 2.9 kg per pack in average and whose surface radiation dose rate ranges from 70 μSv (7 mrem) to 0.85 mSv (85 mrem)/hr.

The average amount of process liquid waste is approximately 0.6 m^3 per cylinder and after treatment its α and β radioactivity becomes 6.7×10^{-3} Bq (1.8×10^{-7} μCi)/ cm^3 (α) and 8.9×10^{-2} Bq (2.4×10^{-6} μCi)/ cm^3 (β), respectively.

3.2 Radiation control

In designing the cleaning facility, much attention was paid to reduce operators' personnel dose rate from cylinder heel adopting various means such as radiation shield and automated precipitate handling. And our experience for three years shows that the results of personnel dose rate is much lower than expected and rather comparable to that of UF_6 conversion area which gives low dose rate. Actual results of dose rate are shown in Table 2.

As for environmental radioactivity, the airborne concentration in the restricted area is below 5% MPC and that of gaseous effluent from the stack is below 0.33% MPC. Concerning the radiation dose rate in the restricted area, it is 0.5 μSv (0.05 mrem)/hr around the roll stands and BG level in the other spots.

The radiation dose rate of the operators is ND (Not Detectable) in both whole body dose and skin dose. At present personnel dose rate is measured every three months as other areas, though in the beginning of operation it was measured every month to have data of dose rate.

4. CONCLUSION

The cleaning and inspection facility has been operated for three years without major problem and a lot of technical data have been obtained. However, there still remain some technical problems to be improved and those to be studied as a matter of future, which are stated below.

- Improvement of waste treatment to increase over-all cleaning capacity
- Increase of inspection capacity
- Uranium recovery from the precipitate
- Study of handling and treatment of waste from heels contaminated by Reprocessed Uranium, which are expected to be handled in near future.

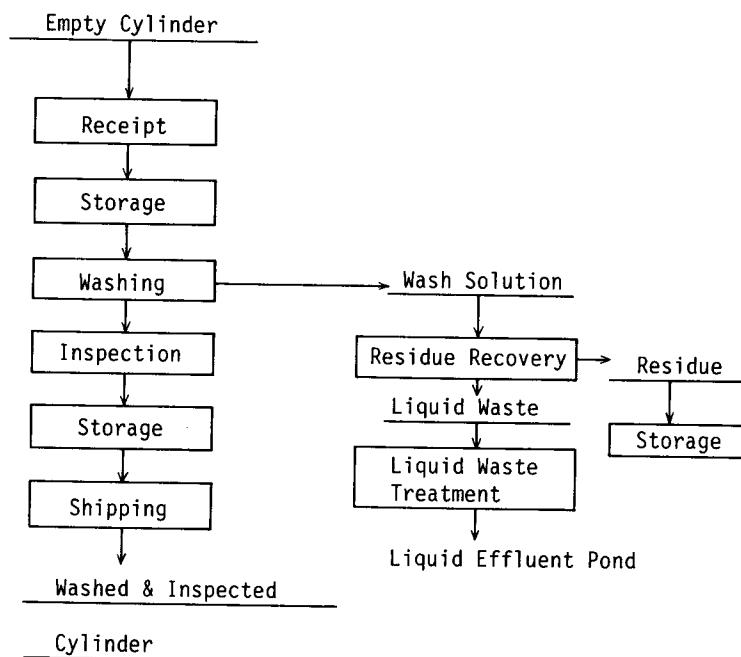


Fig. 1 Total Process

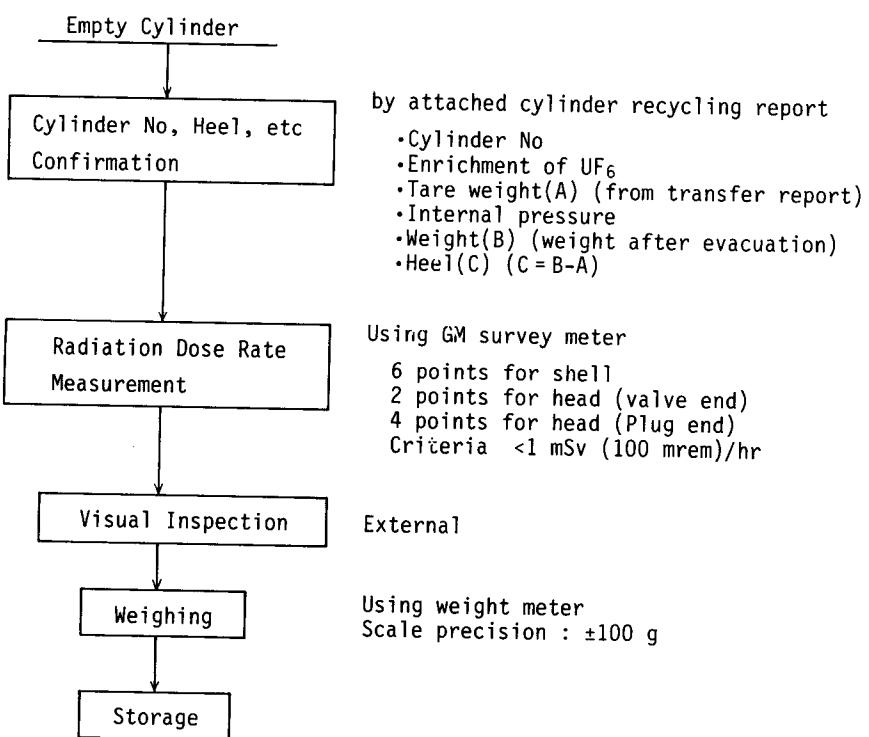


Fig. 2 Receipt and Check Flow Chart

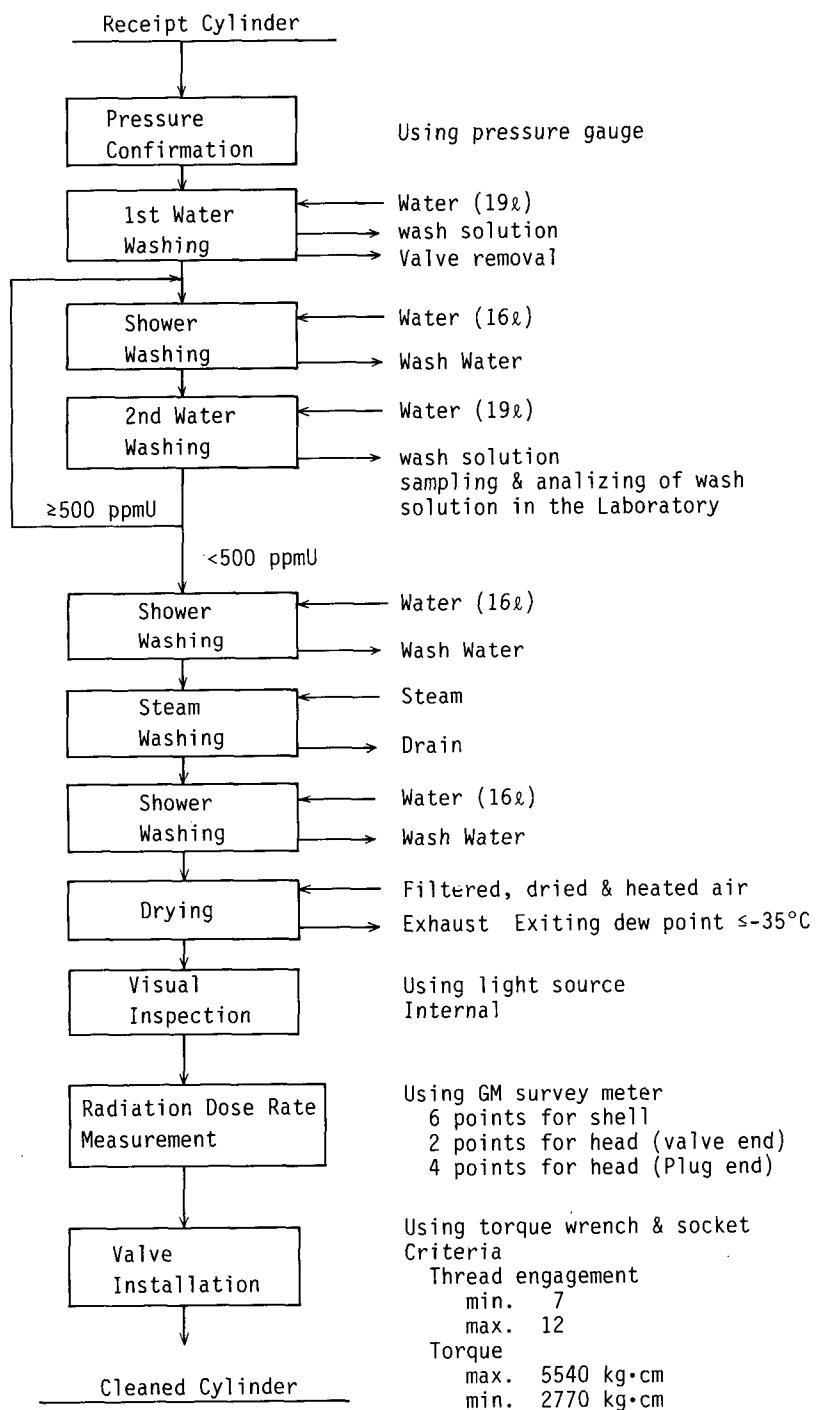


Fig. 3 Cleaning Flow Chart

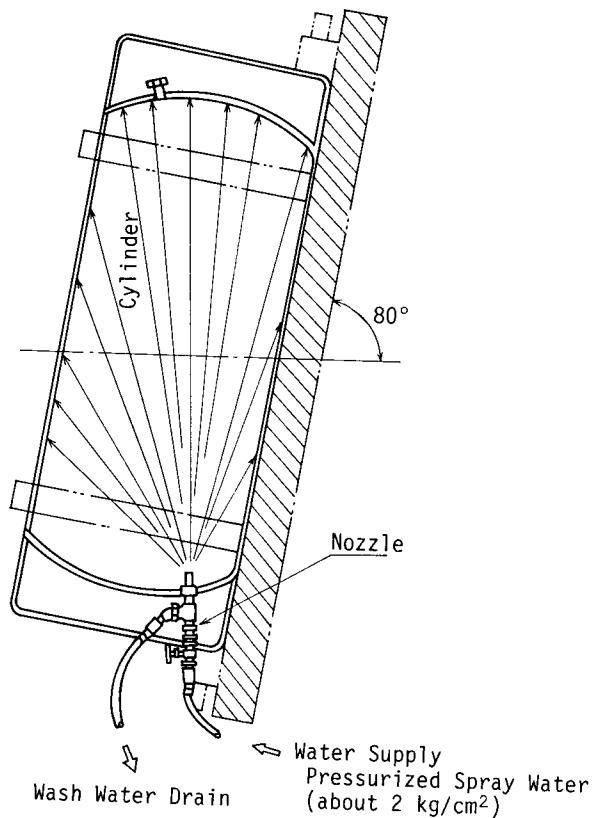


Fig. 4 Shower Washing

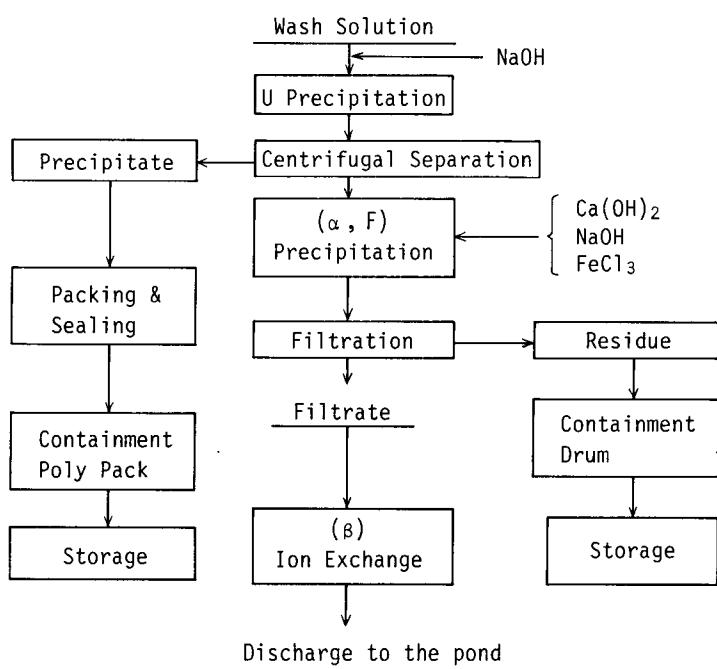


Fig. 5 Treatment of Wash Sioloution

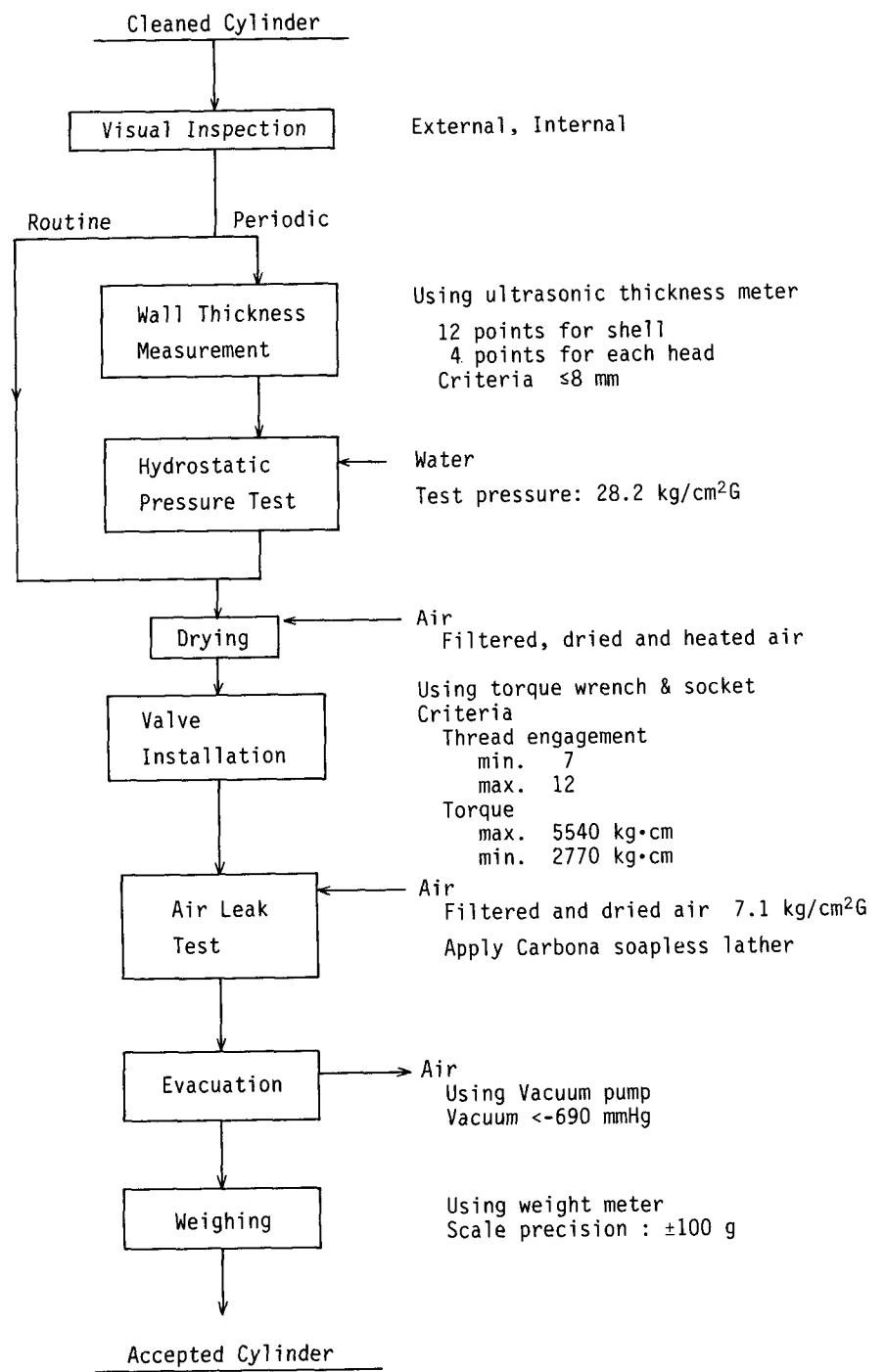


Fig. 6 Inspection Flow Chart

Table 1 Results of Operation

Capacity	1 cyl./shift
Washed Cylinders	340 cylinders (June, 1985 ~ Mar., 1988)
Heel Weight	380 g/cyl. (Ave.) 1971 g (Max.)
Surface Dose Rate (Cylinder)	
• Before Washing	<0.7 mSv (70 mrem)
• After Washing	<1.5 μ Sv (0.15 mrem)
Precipitate	
• Generation Rate	2.6 kg/cyl.
• Packed Weight	2.9 kg/poly. pack
• Surface Dose Rate (pack)	0.07 mSv (7 mrem) ~ 0.085 mSv (8.5 mrem)
Liquid Waste	
• Volume	0.6 m^3 /cyl.
• After Treatment	<ul style="list-style-type: none"> • (a) 6.7×10^{-3} Bq (1.8×10^{-7} μCi)/cm^3 • (b) 8.9×10^{-2} Bq (2.4×10^{-6} μCi)/cm^3

Table 2 Result of Radiation

1.	Airborne Concentration	Regulation
• Restricted Area	$<3.3 \times 10^{-9}$ Bq/ cm^3 ($<0.9 \times 10^{-13}$ μ Ci/ cm^3)	7.4×10^{-7} Bq/ cm^3 2×10^{-11} μ Ci/ cm^3)
• Stack	$<2.5 \times 10^{-10}$ Bq/ cm^3 ($<6.7 \times 10^{-15}$ μ Ci/ cm^3)	7.4×10^{-8} Bq/ cm^3 2×10^{-12} μ Ci/ cm^3)
2.	Radiation Dose Rate	
• Around Roll Stand	0.5 μ Sv (0.05 mrem)/hr	
• Other Area	BG level	
3.	Dose Rate	Regulation
• Whole Body Dose	ND*	0.03 Sv (3 rem)/Quar.
• Skin Dose	ND*	0.08 Sv (8 rem)/Quar.

* ND ; <0.1 mSv (10 mrem)/Quarterly

* Not Detectable

UF6 CYLINDER WASHING AT ANF

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ABSTRACT

Advanced Nuclear Fuels (ANF) Corporation receives UF6 cylinders in its Richland, WA fuel fabrication plant from its utility customers. Following transfer of the UF6, designated cylinders are cleaned by washing using a specialized process which emphasizes personnel safety and low cost. The cleaning operation is conducted in accordance with the guidelines of ANSI Standard N14.1. Specialized equipment has been developed for carrying out this operation with a minimum of space and manpower. Equipment designs and process methods are selected to ensure that the cylinders meet cleanliness requirements while maintaining low personnel radiation exposures, avoiding introduction of organic material and achieving low applied labor. A photo tour will be used to illustrate the equipment and methods used.

INTRODUCTION

Advanced Nuclear Fuels (formerly Exxon Nuclear) has been washing UF6 cylinders for the last 4 years. We have processed about 120 cylinders per year through the wash station. This operation is performed in order to prepare cylinders for periodic inspection and testing to the requirements of ANSI Standard N14.1 Section 6.3, and also to meet requirements of certain overseas customers. Washing is done in accordance with ANSI Standard N14.1, Appendix B. The process outline is shown in Figure 1. Washed cylinders are inspected, dried and sealed. High U wash solution is stored to allow decay of beta-gamma radiation and then processed for uranium recovery. Low U wash solutions are stored in surface impoundments (lagoons) for further uranium recovery and solution disposal. Let us look in more detail at each of these steps.

WASHING

The cylinder is received at the wash station (Figure 2) after it has been weighed, checked to insure it does not exceed a safe batch, and then released by the supervisor (Figure 3). Cylinders with heels exceeding a safe batch are not processed by washing.

The cylinder is placed on a special tilt table for washing and locked into place (Figure 4), and the wash water is added (Figure 5). This table is equipped with a programmable controller which automatically takes the cylinder through a sequence of rolling horizontal and rolling with each end down (Figures 6 & 7). The cycle completes with the cylinder again horizontal and rolling until the operator begins the next step. This allows the operator to continue with other duties during the approximately 30 minutes of the wash sequence.

The wash procedure is as outlined in Appendix B of ANSI N14.1. The variation occurs at the third wash during which is used a solution of 4% ammonia and followed by a wash of 0.2 M HNO3. This is a technique that we have found gives us thorough removal of contaminants. The final wash is with water which is sampled (Figure 8) to verify that the 5000 ppm release limit has been met in the wash solution.

STEAMING & DRYING

A steam lance is inserted into the empty cylinder (Figure 9) and a ventilation "snorkle" is attached for airborne control (Figure 10), directing gasses to a scrubber and HEPA filters. The cylinder is then steamed for one hour. Condensate collects in the rear of the tilted cylinder. As soon as steaming is secured, the cylinder is visually inspected internally (Figure 11) to verify that no UO2F2 deposits remain. The condensate is then pumped out (Figure 12) and air drying is initiated. This is done in fairly rapid sequence to take advantage of the residual heat in the cylinder to aid drying. 600 SCFH of dry, oil-free air is used and the ventilation snorkle is again attached for airborne control (Figure 13). The cylinder is again visually inspected (Figure 14) for cleanliness and dryness. If any oil contamination is suspected, a Freon wash is used. Vacuum cleaning is used for removal of loose scale or other solids (Figure 15).

CYLINDER SEALING

When all cleaning steps are completed and the cylinder cleanliness certified by the supervisor (Figure 16), the cylinder is sealed. For those cylinders designated for periodic inspection and testing under ANSI N14.1 Section 6.3 a 1 inch pipe plug is installed. This is later replaced by a special valve assembly at the hydrostatic test station. For other cylinders, a cylinder valve is installed (Figures 17 & 18) and leak checked at 100 psi. Certain European customers request that cylinders be pressurized with nitrogen to 5 psig. For other destinations, the cylinder is evacuated to 10 in. Hg (Figure 19). The cylinder is then ready for removal from the station and survey by Health Physics for release from the building (Figure 20). Total cycle time is 24 hours with 4 hours applied labor.

LIQUID PROCESSING

In light water reactor fuel fabrication we normally encounter only alpha radiation sources. However, the vaporization process tends to leave behind less volatile decay products such as Thorium. This results in requirements for special radiation monitoring instrumentation at the wash station and a decay period being used on all wash solutions prior to further processing.

The first two wash solutions contain the majority of the uranium as well as the high beta/gamma contaminants. These solutions are pumped to a holding tank (Figure 21) and then, when washes from 4 cylinders have been accumulated, transferred to a plastic 55-gallon drum (Figure 22). Drums, averaging 6.5 kg U each, are stored for at least 6 months to allow daughter product decay. Following the storage period, solution from the drums is pumped back into the process area for recovery by ADU precipitation, purification by solvent extraction and return to working stock for subsequent fabrication.

Wash solutions after the first two rinses are pumped to a storage lagoon (Figure 23) where they are combined with other waste from the production area. These are eventually processed for uranium and ammonia recovery and then discharged.

CONCLUSION

We have seen the equipment and processes used at ANF for washing UF₆ cylinders. The final product of this process is cylinders meeting the cleanliness requirements of ANSI N14.1. We are able to recover the full economic value of the heels and we are able to achieve this in a safe and economical fashion.

REFERENCE:

ANSI Standard N14.1-1982, American National Standard for Packaging of Uranium Hexafluoride for Transport.

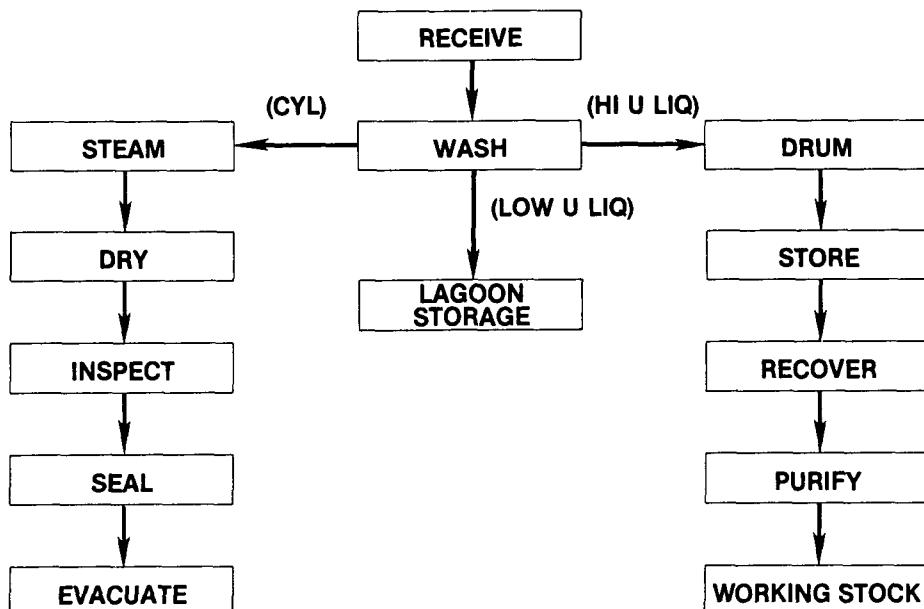


Figure 1

UF₆ Cylinder Washing at ANF
Process Outline

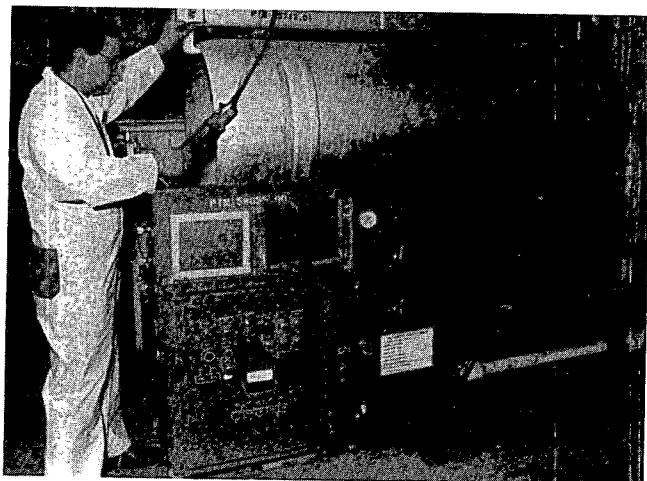


Figure 2
Cylinder Placed on Wash Station

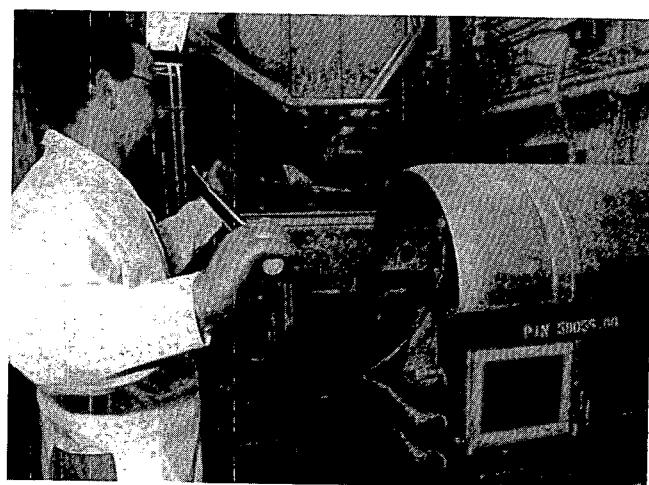


Figure 5
First Wash Introduced

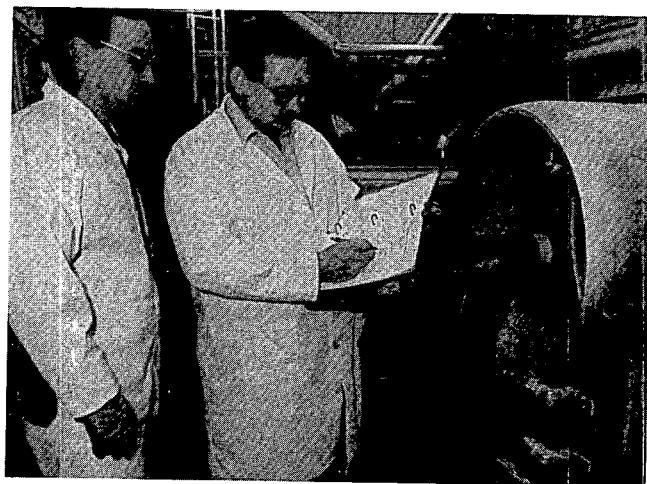


Figure 3
Supervisor Release for Washing

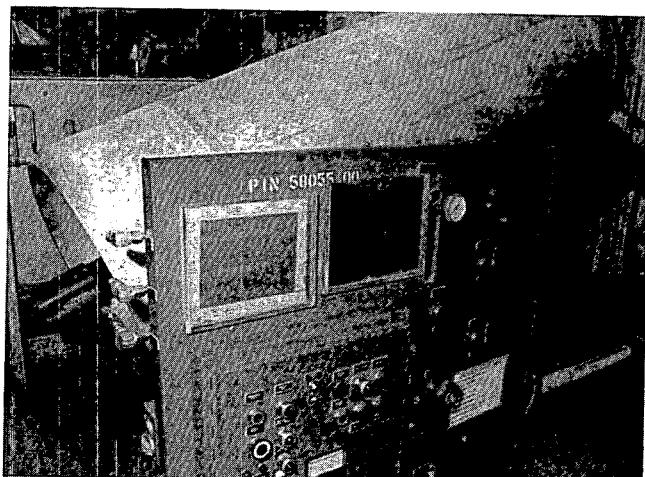


Figure 6
Tilt Table In Operation



Figure 4
End Stop Locked

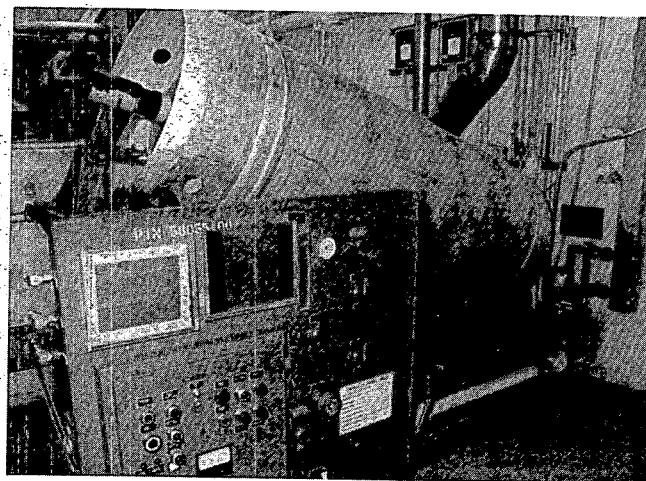


Figure 7
Tilt Table In Operation



Figure 8
Wash Solution Sampled



Figure 11
Inspection Light Inserted



Figure 9
Steam Lance Inserted

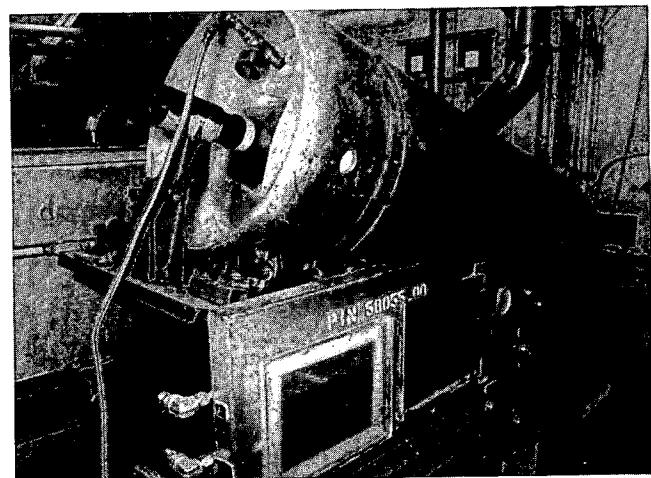


Figure 12
Condensate Pumped Out



Figure 10
Exhaust Snorkle Attached

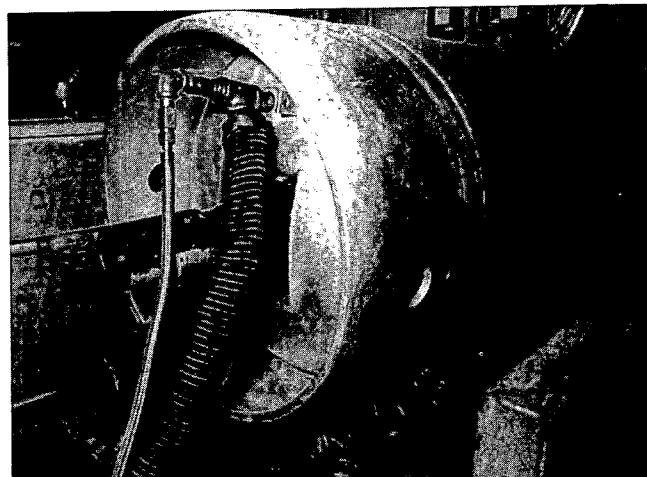


Figure 13
Air Drying

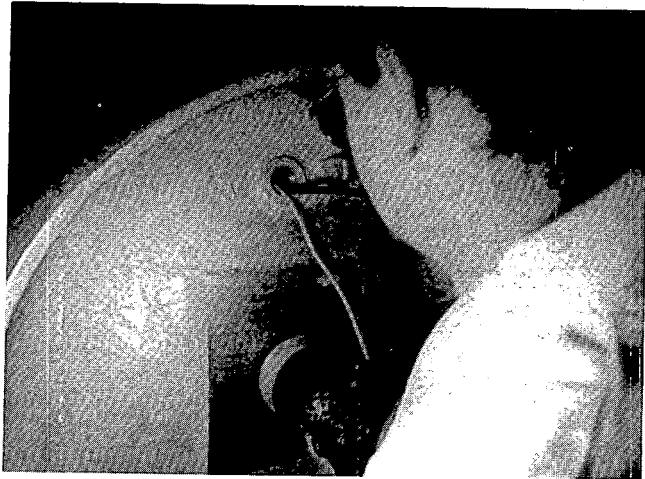


Figure 14
Visual Inspection



Figure 17
Cylinder Valve Installation

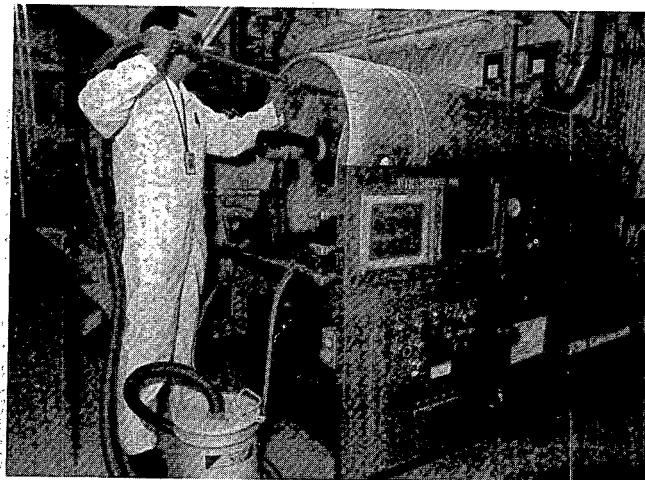


Figure 15
Vacuum Cleaning

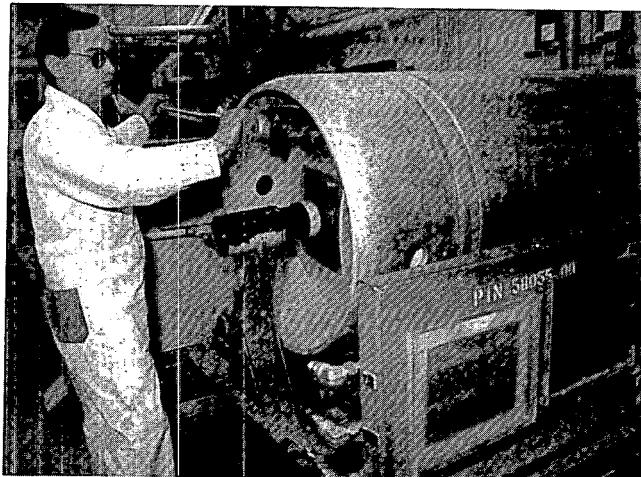


Figure 18
Cylinder Valve Torquing

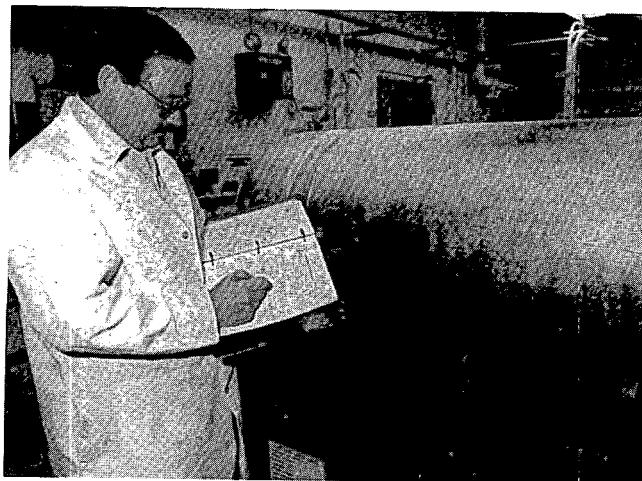


Figure 16
Supervisor Release of Clean Cylinder

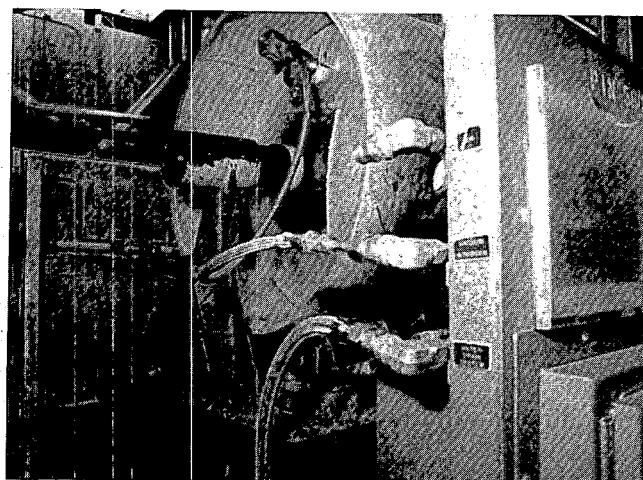


Figure 19
Evacuation

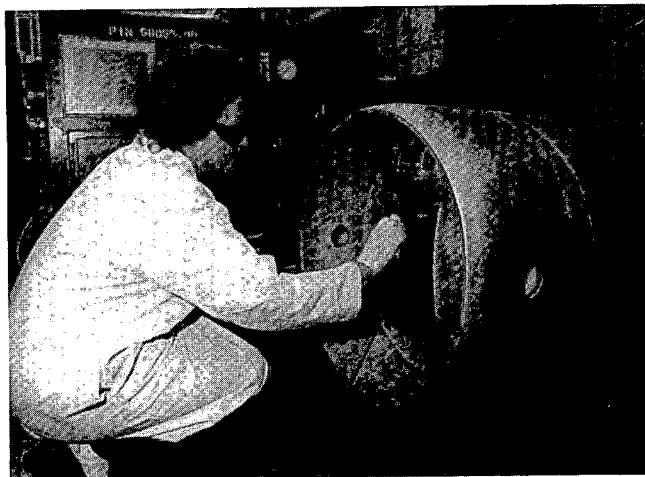


Figure 20
Radiation Survey and Release



Figure 22
Drums for Wash Solution Aging

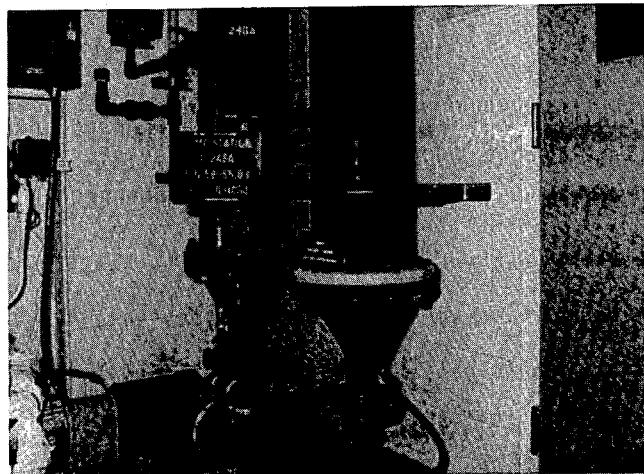


Figure 21
Wash Solution Holding Tanks



Figure 23
Liquid Waste Storage Lagoons

UF₆ CYLINDER INSPECTION AND TEST FACILITY AT PADUCAH GASEOUS DIFFUSION PLANT

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ABSTRACT

Paducah cylinder washing and testing facility operations are described for the cleaning of 10- and 14-ton uranium hexafluoride (UF₆) cylinders. Decontamination solution preparation and maximum reuse of solution are explained. Cylinder inspection parameters before and after cleaning a cylinder are detailed on data tables. Cleaning procedures for a 10- and 14-ton cylinder are outlined and the equipment is shown in photographs and process diagrams. A 12-point radiation survey of the cylinder before and after cleaning is used in protecting the worker and maintaining exposure to radiation as low as reasonably achievable (ALARA). The inspection of UF₆ cylinders is required for new, in-service, and damaged cylinders as detailed in the criteria in ANSI N14.1 and Oak Ridge Operation ORO-651. Criteria and inspection procedures for the replacement of 1-inch UF₆ cylinder valves and 1-inch and 1 1/2-inch cylinder plugs and the parameters for thread engagement of valves and plugs are outlined. A uniform method of measurement for thread insertion made on both valves and plugs is described. Specialized equipment to facilitate installation of the plug and valve and reduce worker exposure to undue hazards are described. Hydrostatic testing of cylinders, drying, and evacuation are part of the Chemical Operations Department's function and they are presented in the paper. Also discussed is the refurbishment of 1-inch UF₆ cylinder valves.

INTRODUCTION

Paducah cylinder inspection and testing facility operations are described for 10- and 14-ton UF₆ cylinders. The cylinder wash and test facility is located in the Chemical Operations building at the Paducah Gaseous Diffusion Plant (PGDP). The system is designed as an always-safe unit for handling uranium up to 2.0 wt % U²³⁵.

Prepared by the Paducah Gaseous Diffusion Plant, Paducah, Kentucky 42001, operated by MARTIN MARIETTA ENERGY SYSTEMS, INCORPORATED, for the U.S. DEPARTMENT OF ENERGY, under Contract No. DE-AC05-84OR21400.

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The cylinder washing system consists of two 150-gallon tanks, a solution spray and transfer pump, a drain pan of proper slab thickness for criticality control, spray nozzles, and evacuation connections. An air-operated Milton Roy metering pump is used to obtain and maintain the required hydrostatic test pressure. The drying system consists of an electrically heated enclosure, a dry air purge connection, dew point meters, and an evacuation jet.

DECONTAMINATION SOLUTION PREPARATION

The decontamination agent used in cylinder cleaning is sodium carbonate. Decontamination solution preparation is accomplished by adding 200 gallons of water to the east and west solution storage tanks and 100 pounds of sodium carbonate. The pH should be basic (9-11) and will be one of the parameters monitored to determine when the solution should be rejuvenated. If the pH goes below 5, another 100 pounds of sodium carbonate should be added to the solution. The pH is checked in the solution before and after each cylinder is washed. If ten cylinders have been cleaned since the last addition of soda ash, then another 100 pounds of sodium carbonate is added unless the solution's pH is over 9. If the addition of the 100 pounds of sodium carbonate does not bring the pH up above 7, then the solution is ready to be changed. Another indication of the level of contamination is the pressure gage reading from the spray pump when circulating the cylinder cleaning solution. The pressure of the clean solution is 60- to 70-pound gage. Pressure of a dirty solution ready to be transferred is 100- to 110-pound gage. Crystal formation in the pan is another sign that transfer of solution may be required.

CLEANING PROCEDURES FOR A UF₆ CYLINDER

A physical inspection of the cylinder to be washed is made to ensure that it has not been damaged. Nameplates which are badly corroded must be replaced to ensure legibility. Also, the loose rust is cleaned from the cylinder before placing it on the turning fixture to minimize contamination of the solution with iron.

Before and after washing the cylinder, dry surface radiation readings must be taken in 12 different

locations. The Victoreen 491 survey meter (shield open) should be used to detect the gross gamma-beta activity. A reading of 1 mR/hr, or greater, indicates a cylinder which has not decayed sufficiently. The last feed data of this cylinder should be verified before proceeding to wash.

The cylinder to be washed is placed in the cradle on the turning fixture. The cylinder tie-down cables are tightened securely with the turn-buckles. Check the cylinder for vacuum by attaching the compound gage fixture to the cylinder valve and opening the cylinder valve.

If the cylinder has a vacuum, a special adapter which consists of spray nozzle, compound pressure gage, and valve has been fabricated to screw into the plug hole. The plug from the cylinder is removed and replaced by the special adapter. The solution valves and the special adapter valve are opened to spray for five minutes. If no pressure is evident from the compound pressure gage reading, then the special adapter is removed.

Cylinders at slight pressure are relieved by connecting the water spray nozzle to the cylinder valve and operating for three minutes. The spray nozzle is disconnected from the cylinder valve and the evacuation valve is connected to the cylinder valve and both valves then opened. This will relieve any pressure. After reducing the pressure, the evacuation valve is disconnected and the cylinder valve removed.

After equalizing any pressure or checking for vacuum, the drain adapter with a quick-opening valve is screwed into the plug hole. The cylinder is rotated and positioned for complete draining. The drain pan ejector suction valve and the quick-opening valve on the drain adapter in the cylinder are opened. The cylinder is raised to the full vertical position after draining. The spraying operation is repeated two or three times. The final water spray nozzle is inserted into the valve hole and the water valves opened. The cylinder interior is sprayed until the drain solution is clear (approximately one minute). An inspection light is inserted through the plug opening and the cylinder inspected for cleanliness through the valve opening. A flow diagram of the system is shown in Figure 1.

HYDROSTATIC TESTING OF CYLINDERS

Cylinder valves must be installed with the proper thread engagement, as well as the proper torque, which is between 200 and 400 ft-lb. Table 1 shows the minimum and maximum number of threads showing for acceptance of a valve in a UF₆ cylinder. The use of teflon tape or oil on the threads is prohibited.

Cylinder plugs must be installed with the proper thread engagement which is a maximum of four visible threads and a minimum of one visible thread. The UF₆ cylinder plug thread insertion is measured by the following formula:

L = Length of plug

PH = Thickness of plug head

TV = Threads visible

H = Shoulder of plug above the threads and below the head

L-PH-SH = Thread length of plug (TLP)
TLP/.087 = Number of threads on plug (NTP)
NTP - TV = Number of threads engaged in coupling (TE)

The threads engaged (TE) must be a minimum of five and maximum of eight. The plug torque should be between 200 and 650 ft-lb.

All torque readings and the threads visible readings are logged. The torque wrench must be certified on a quarterly basis or as needed. The use a torque wrench which has an expired certification date is prohibited.

A new valve is installed in the cylinder and opened. The cylinder is rotated to a near-vertical position. A high pressure pigtail from the water line is connected to the cylinder plug opening. The cylinder is positioned to allow all air to be bled from the cylinder when filled with water for hydrostatic testing. The inlet water valves are opened and the cylinder filled until water flows out of the cylinder valve. The air pressure regulator is set to obtain the desired hydrostatic pressure. When the desired hydrostatic pressure in the cylinder is obtained, the Quality Evaluation Department personnel observe and certify the testing operation. An air hose is connected to the cylinder valve and the air valve opened to help drain the water from the cylinder. The cylinder is then ready to be moved to the cylinder drying station.

All DOE-owned cylinders and all customer-owned cylinders fabricated in the United States are code stamped. DOE has reviewed all non-USA-made UF₆ cylinders to verify that they are manufactured in accordance with ANSI N14.1 specifications, which does include code stamping. Foreign countries do not use the U-symbol, which is an ASME requirement, to indicate code stamping. However, the hydrostatic test date on these non-USA-made UF₆ cylinders will expire under the same conditions as those cylinders manufactured in the United States.

The hydro is done at two times the maximum absolute working pressure. The hydrostatic pressure test date expires five years after the year of manufacture and five years after the year in which any subsequent pressure retest occurs. These yearly dates are stamped into the cylinder nameplate. For example: the hydrostatic pressure test date on a cylinder dated 1981 will expire at the end of 1986.

CYLINDER DRYING AND EVACUATION

The cylinder is set in the drying station and the pigtailed connected to the plug opening and the cylinder valve. The radiant heaters are energized with the temperature controller set at 200°F. The radiant heaters will not operate unless the flow indication on the flow interlock gage is at least 40 divisions. After two hours drying time, the dew point of the exhaust air from the cylinder is checked. A sample stream is directed to the Panametrics Model 700D Hygrometer (dew point meter). The dew point should be -30°F, or less. If the dew point is -30°F, the block valve is opened to relieve the pressure. If the dew point is not -30°F, then further drying is necessary.

The pigtail is removed from the plug end and a plug installed in the cylinder. The cylinder is pressurized to 80 psig and the area around the plug and the valve soap tested. When the cylinder is airtight, the pressure is relieved. If it is necessary to tighten the plug or valve, the cylinder is evacuated to 20 inches Hg. The cylinder is removed from the drying stand and the official new tare weight established.

SOLUTION TRANSFER FROM THE WASHING SYSTEM

The wash solution is sampled for assay. The solution is agitated thoroughly before sampling. If the assay is less than 1.0 wt % U^{235} , the solution is safe to transfer to the storage tanks and onto the dissolvers. If the assay is >1.5 wt % U^{235} , the solution is isotopically diluted by washing cylinders of lower assay. After verification of the assay and valving to the acidifying tanks, the solution is transferred, after it has heated to 120°F, to the C-400 recovery system. Three hundred gallons are transferred to the two storage tanks and 150 gallons for flushing out the drain pan and the lines to the acidifying tanks.

UF₆ CYLINDER INSPECTION

An inspection is to be performed on any UF₆ cylinder prior to shipment from the plant or immediately after receipt into the plant. UF₆ cylinders covered include 10- and 14-ton cylinders. Documentation of the results of this inspection is to be made. The records of the inspection of any UF₆ involved in any off-site movement on the UF₆ Cylinder Inspection Data Sheet (UCN-9009) are recorded. A copy of this form is shown in Figure 2. Procedures and standards under which UF₆ cylinders are inspected, include ASME, ANSI N14.1, Oak Ridge Operation ORO-651, PGDP Quality Evaluation (QE) SOPs QE-103 and QE-108. Cylinders are procured from qualified vendors. Vendor audit and surveillance are performed periodically to assure adherence to specifications. All new cylinders are inspected 100% visually. One cylinder per shipment is tested and inspected internally 100% upon receipt.

Any cylinder which is found to be unacceptable is immediately tagged. Cylinders which are found to be damaged, overfilled, or to have a defective valve are to be tagged with a Defective Cylinder Tag (UCN-11300). Code stamped cylinders involved in off-site shipments, which are found to have an expired hydrostatic pressure test date, are to be tagged with a Hydrostatic Pressure Test Date Expired Tag (UCN-11301).

Shipment of cylinders is withheld from the carrier when any unacceptable damage or condition is found. A customer-owned cylinder may be shipped off-site if the hydrostatic pressure test date is expired, providing it has a Hydrostatic Pressure Test Date Expired Tag attached to the valve.

The Quality Evaluation Department will determine when a cylinder is found to have any unacceptable damage as indicated in Figure 3. The Quality Evaluation Department will make the final decision as to whether the cylinder is acceptable or unacceptable and will record their decision by completing Section A at the bottom of Form

UCN-9009. Any qualifications for the use of cylinders which are approved for limited use (until the defect(s) is corrected) must be entered in Section A of Form UCN-9009.

It must be verified that all full cylinders have been allowed to cool until the UF₆ contents have solidified. The minimum cooling period for the solidification of the UF₆ contained in full cylinders is five days for either 10- or 14-ton cylinders.

A cold-pressure check is performed on the cylinder to be shipped using a compound pressure gage and cylinder valve adapter. All full or empty cylinders being shipped from the Paducah Plant are to be cold-pressure checked at ambient temperature to assure that the internal cylinder pressure is below atmosphere.

A tamper-proof device is installed over the cylinder valve and a numbered seal affixed to the tamper-proof device.

PROCEDURE FOR REUSE OF CONTAMINATED ONE-INCH UF₆ CYLINDER VALVES

Section 6.3.3 of ANSI N14.1-1982, American National Standard for Packaging of Uranium Hexafluoride, permits the reuse of UF₆ cylinder valves provided they are carefully inspected and tested to verify their suitability for continued use. Each cylinder valve is completely disassembled. The outlet port cap gasket and the stem packing are discarded. Each valve component is closely examined for obvious damage. The heat identification number stamped on each valve body is examined and those valve bodies having a heat identification number of 16, 17, 18, 19, 20, 21, or 22 are segregated. Using a .835-inch diameter plug gage, the bore of the inlet port of each cylinder valve body is checked to verify that it is not excessively necked-down. The minimum bore diameter, anywhere along its approximate 1.25-inch length, should not be less than 0.835 inches. All packing nuts are transported to the C-720 Machine Shop area for stress relieving. The stress relief operation consists of the packing nuts attaining a temperature between 700 and 720°F and being held at the temperature for a minimum of ninety minutes. All valve components are decontaminated, including the packing nuts which have been stress relieved. Each set of valve components are reassembled per drawing E-J-11246, Revision 15. One or two drops of fluorinated lubricant are applied to the Acme threads of each valve stem, and to the threads of the packing nut, prior to their assembly. A 1/8-inch high letter "R" is stamped immediately below the alloy number 636 on each valve body to designate that the valve has been refurbished. Maintenance Division personnel shall tin the threads of each cylinder valve inlet port.

The Quality Evaluation Department is contacted and requested to witness and certify the high pressure leak tests. The outlet port cap on each valve outlet port is assembled with 50 ft-lbs torque and installed in each valve at the high pressure leak test fixture. The entire valve is submerged in a test tank of water, the valve body is pressurized to 400 psig, and observed for leakage around the area where the stem exits the packing, around the

packing nut threads, and around the outlet port cap threads. With the valve seated at 55 ft-lbs torque, again the entire valve and test fixture are submerged in the test tank of water and pressure tested for seat leakage at 400 psig. If no leaks, the valve is dried and an ACCEPTED sticker is dated using indelible ink and applied to each valve by QE Department personnel.

Table 1

Type of Cylinder	Valve Size	Type of Valve	Acceptable No. of Threads Showing	
			Min.	Max.
MD				
Model 12A	3/4"	SUPERIOR 5665*	4	9
Model 12B	3/4"	SUPERIOR 5665*	4	9
2-1/2 TON				
Model 30A without coupling	3/4"	SUPERIOR 5665*	4	9
Model 30A without coupling	1"	SUPERIOR-UNMODIFIED	1	10
Model 30A without coupling	1"	MODIFIED**	1	7
Model 30A with coupling	1"	SUPERIOR-UNMODIFIED	1	10
Model 30A with coupling	1"	MODIFIED**	1	7
Model 30B	1"	SUPERIOR 11246	1	7
10- and 14-TON				
Thick-wall without coupling	1"	SUPERIOR-UNMODIFIED	5***	10
Thick-wall without coupling	1"	MODIFIED**	5***	7
Thick-wall with coupling	1"	SUPERIOR-UNMODIFIED	1	10
Thick-wall with coupling	1"	MODIFIED**	1	7
Thin-wall	1"	SUPERIOR-UNMODIFIED	1	10
Thin-wall	1"	MODIFIED**	1	7

*Or approved equal.

**Superior 11246 or approved equal.

***Five threads showing is required to provide room to connect the pigtail to the cylinder valve.

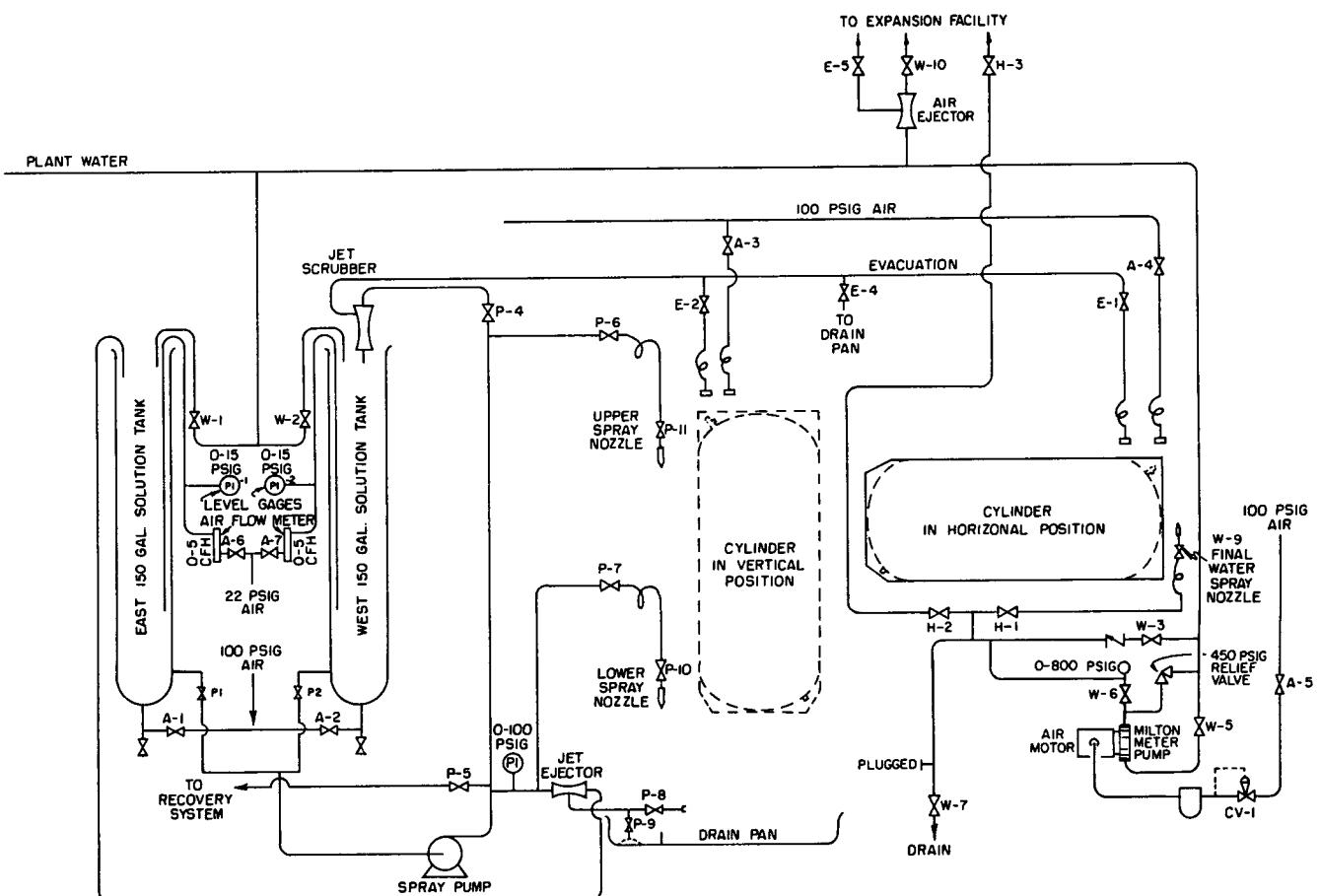


Figure 1. Schematic Diagram of the Spray System for the Cylinder Cleaning and Test Facility in Building C-400

UF₆ CYLINDER INSPECTION DATA SHEET

CYLINDER NUMBER <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <input type="checkbox"/> 30A (2½-ton) <input type="checkbox"/> 48F (14-ton HW) <input type="checkbox"/> <input type="checkbox"/> 30B (2½-ton) <input type="checkbox"/> 48Y (14-ton HW) <input type="checkbox"/> <input type="checkbox"/> 48A (10-ton) <input type="checkbox"/> 48G (14-ton LW) <input type="checkbox"/> <input type="checkbox"/> 48X (10-ton) <input type="checkbox"/> 48H (14-ton LW) <input type="checkbox"/> </div> <div style="width: 45%;"> <input type="checkbox"/> Date Shipped <input type="checkbox"/> Date Received </div> </div> <p>Cylinder is Code Stamped <input type="checkbox"/> No <input type="checkbox"/> Yes</p> <p>CYLINDER STATUS <input type="checkbox"/> Full <input type="checkbox"/> Empty</p>		CYLINDER MODEL <p>Hydrostatic Pressure Test Date of _____</p> <p>is <input type="checkbox"/> Acceptable <input type="checkbox"/> Not Acceptable</p> <p>CYLINDER'S CONTENTS ARE SOLIDIFIED <input type="checkbox"/> Yes <input type="checkbox"/> No</p> <p>CYLINDER BEING INSPECTED <input type="checkbox"/> Prior to being shipped <input type="checkbox"/> After being received <input type="checkbox"/> Prior to being filled <input type="checkbox"/> Prior to being heated</p>																			
<p>Cylinder Is Overfilled: <input type="checkbox"/> No <input type="checkbox"/> Yes, Net Weight is _____ Pounds; Maximum Allowable Fill Limit is _____ pounds.</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 15%;"></th> <th style="width: 85%; text-align: center;">CONDITION</th> </tr> <tr> <th></th> <th style="text-align: center;">Acceptable Un-acceptable Not Applicable</th> </tr> </thead> <tbody> <tr> <td style="vertical-align: top;">I. CYLINDER VALVE, VALVE PORT AND PLUGS</td> <td> <p>A. VALVE: 1. Valve Type _____ 2. Physical Damage _____ 3. Thread Engagement _____ 4. Valve Cap - Present and in Place _____</p> <p>B. VALVE PORT: 1. Plugged with UF₆ _____ 2. Contaminated with Other U-Salts or Foreign Material _____</p> <p>C. PLUGS: 1. Physical Damage _____ 2. Thread Engagement _____ 3. Sealed _____</p> <p>D. VALVE PROTECTOR: 1. Present and Properly Positioned _____ 2. Sealed _____</p> <p>Description of Damage (if any): _____</p> </td> </tr> <tr> <td style="vertical-align: top;">II. CYLINDER WELDS</td> <td> <p>A. CIRCUMFERENTIAL HEAD SEAM WELD - VALVE END _____ B. CIRCUMFERENTIAL HEAD SEAM WELD - PLUG END _____ C. LONGITUDINAL SEAM WELD _____</p> <p>Description of Damage (if any): _____</p> </td> </tr> <tr> <td style="vertical-align: top;">III. CYLINDER SHEEL AND HEADS</td> <td> <p>A. SHELL _____ B. HEAD - VALVE END _____ C. HEAD - PLUG END _____</p> <p>Description of Damage (if any): _____</p> </td> </tr> <tr> <td style="vertical-align: top;">IV. STIFFENING RINGS</td> <td> <p>A. VALVE END _____ B. CENTER _____ C. PLUG END _____</p> <p>Description of Damage (if any): _____</p> </td> </tr> <tr> <td style="vertical-align: top;">V. SKIRTS</td> <td> <p>A. VALVE END _____ B. PLUG END _____</p> <p>Description of Damage (if any): _____</p> </td> </tr> </tbody> </table>					CONDITION		Acceptable Un-acceptable Not Applicable	I. CYLINDER VALVE, VALVE PORT AND PLUGS	<p>A. VALVE: 1. Valve Type _____ 2. Physical Damage _____ 3. Thread Engagement _____ 4. Valve Cap - Present and in Place _____</p> <p>B. VALVE PORT: 1. Plugged with UF₆ _____ 2. Contaminated with Other U-Salts or Foreign Material _____</p> <p>C. PLUGS: 1. Physical Damage _____ 2. Thread Engagement _____ 3. Sealed _____</p> <p>D. VALVE PROTECTOR: 1. Present and Properly Positioned _____ 2. Sealed _____</p> <p>Description of Damage (if any): _____</p>	II. CYLINDER WELDS	<p>A. CIRCUMFERENTIAL HEAD SEAM WELD - VALVE END _____ B. CIRCUMFERENTIAL HEAD SEAM WELD - PLUG END _____ C. LONGITUDINAL SEAM WELD _____</p> <p>Description of Damage (if any): _____</p>	III. CYLINDER SHEEL AND HEADS	<p>A. SHELL _____ B. HEAD - VALVE END _____ C. HEAD - PLUG END _____</p> <p>Description of Damage (if any): _____</p>	IV. STIFFENING RINGS	<p>A. VALVE END _____ B. CENTER _____ C. PLUG END _____</p> <p>Description of Damage (if any): _____</p>	V. SKIRTS	<p>A. VALVE END _____ B. PLUG END _____</p> <p>Description of Damage (if any): _____</p>				
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DISTRIBUTION: White - Uranium Control (KYRC) Blue - Quality Evaluation (When Section A is Completed) Buff - Originator <small>UCN-9009 (5 6-86)</small>		CONDITIONED LEGEND: A - Acceptable B - Unacceptable NA - Not Applicable																			

Figure 2. Typical Cylinder Inspection Data Sheet

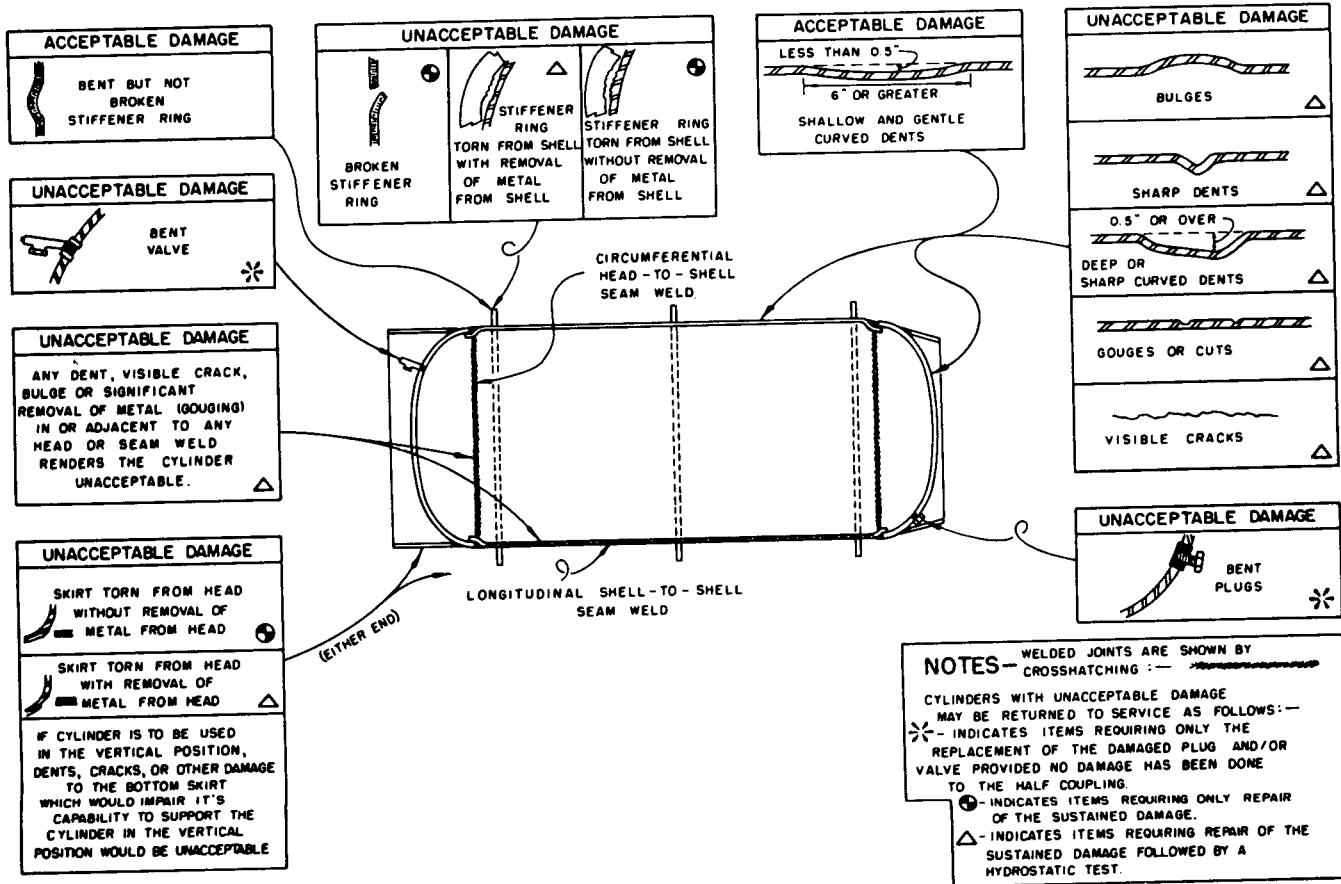


Figure 3. Examples of Acceptable and Unacceptable Damage to UF_6 Cylinders

Mr Guy LAMORLETTE

Mr Jean-Paul BERTRAND

1- TRICASTIN : AN INTEGRATED SITE GEARED TO THE NEEDS OF THE UF6 INDUSTRY

The French nuclear power program is characterized by a high level of standardization and by the streamlining of production methods. The following organizations are responsible for the program's development :

- . EDF, a publicly owned industrial group responsible for the production and distribution of electricity.
- . Government-owned corporations, such as COGEMA which is responsible for the nuclear fuel cycle (fig 1).
- . The CEA - French Atomic Energy Commission - the national research arm of the program, which provides the government with technical studies and programs for safety control and monitoring.

With the exception of uranium mining, activities of the front-end of the fuel cycle are concentrated right on the Tricastin site.

Some of these companies are engaged in bilateral activities with foreign companies, but they are all engaged in a contractual relationship with COGEMA.

In this framework, COGEMA's Pierrelatte facility, especially its Uranium Management Facility (UMF) acts as a sort of revolving door, coordinating the various on-going industrial activities at the site (fig 2).

. Administration of inventory equipment to adequately ensure the flow of UF6 entering and existing the plant's facilities.

. Quality control of the UF6 product from natural uranium to enriched uranium, including UF6 from reprocessing.

. Containers maintenance.

. Implementation and planning of UF6 shipments throughout the world, including bookkeeping of uranium contracts.

These various activities are detailed further on, with numbers illustrating the activities that took place in 1987.

2 - UF6 HANDLING AND STORAGE

2.1 Natural :

Deliveries of natural UF6 are the responsibility of COMURHEX and occur on a regular basis. Deliveries are subject to qualitative and quantitative controls by COGEMA. These controls are performed either on a systematic basis, especially with regard to the quality of packaging, or on a more random basis to control the quality of the material itself.

The power requirements of the EURODIF enrichment plant vary throughout the year in a one-to-three ratio. In France, at winter time, the demand for electric heat is high, so that, at that time, EURODIF lowers its energy consumption. As a result, the need for natural uranium also decreases. On the summertime, EDF is in a better position to provide EURODIF with lower-cost electricity for the production of enrichment services in larger batches : UF6 used can reach 1,000 metric tonnes U/month.

This seasonal adjustment led to the creation of a intermediary inventory set up between COMURHEX and EURODIF. The uranium hexafluoride storage area at the UMF can accomodate up to 6,000 UF6 containers, including many containers containing natural UF6.

The natural UF6 containers are reutilized after being drained, with their UF6 heels for material to be filled up again at the convertor.

In the context of COGEMA's enrichment services sales to its foreign customers, the UMF is also sent natural UF6 from the world's convertors. If a quick return of the containers is required by the convertor, they are exempted from the standard waiting procedure.

2.2 Enriched :

Most of the enriched UF6 delivered to the UMF originates in EURODIF. Transportation on site allows us not to use the protective overpacks between the EURODIF plant and the UMF, and even if necessary, to the FBFC fuel fabrication plant. This allows for greater flexibility and reliability with full guarantees for the security and physical protection of the materials.

UMF storage can be divided into two distinct categories :

- An intermediary stage between the enricher and fuel fabricators, similar to the inventory account of natural uranium. UMF deliverers enriched uranium to FBFC, but also, on behalf of COGEMA's customers, to the world's fuel fabricators.
- An inventory account to be used to respond to last minute changes in customer orders, that are occurring more and more frequently. To this end, a facility for the isotopic adjustment of UF6 to a liquid or gaseous phase, is used. Programs are optimized to reduce enrichment losses that result from these mixtures.

The quality of enriched uranium is scrupulously controlled at UMF. Prior to any delivery to a fuel fabricator, the UF6 undergoes checks according to French and international specifications. In particular, an independent company systematically verifies on behalf of its customers that UF6 conditioning and sampling of enriched UF6 follow the US NRC norms.

After the enriched UF6 containers are emptied, they are delivered back to the UMF. In general, all the UF6 in the containers has, by this time, been taken out by the fuel fabricator. This is always the case in Europe.

In the United States, conditions vary, since heels often remain in emptied containers.

COGEMA believes that this can lead to lasting problems for operators and customers including :

- . A future shipment could be contaminated by the remaining heels.
- . Difficulties tied to increased transportation costs in the case where rules would require shipment in overpacks.
- . More complex material accountability.

We could hope that US fuel fabricators would adopt the practice of returning fully emptied containers.

2.3 Depleted uranium :

It is not possible to keep in inventory the totality of the depleted uranium from the EURODIF plant, because of environmental requirements.

Depleted uranium is taken over by the UMF and is temporarily stored before being transformed into stable oxide for long-term storage. Transfer of UF6 between the depleted product from the EURODIF plant and the defluorination plant on the COGEMA Pierrelatte site occurs at a rate of 10,000 metric tonnes/year. The defluorination plant transforms UF6 into very low reactivity U3O8.

At the plant's entrance, isotopic controls are made with a CIND, an instrument developed by COGEMA and commercialized at the AIEA, among others.

The recovered hydrofluoric acid is extremely pure and can be used in fine chemistry. COGEMA is thus among the leading European producers of liquid hydrofluoric acid.

2.4 Reprocessed Uranium (REPU)

To this date, UF6 from reprocessing is not produced in very large quantities.

In the future, following the start-up of COGEMA's UP3, UP2-800 reprocessing plants as well as Sellafield plant, quantities of reprocessed uranium available in Europe will rapidly increase.

It is up to the operators of facilities such as the UMF, to make sure that uranium from reprocessing is kept separate from the materials to limit the production of artificial uranium isotopes (U236) :

- . multiple conditioning lines must be installed, or else carefully swept and purged.
- . UF6 from reprocessing must be conditioned in especially selected containers. Internal clean-up of these containers produce special effluents that are treated in a process carefully managed by COGEMA.

3 - UF6 QUALITY CONTROL

A high level of quality can be achieved if at least three conditions are met :

- . Follow clear, realistic, albeit constraining, criteria that are the result of a track record with UF6 accumulated over the past 40 years in the UNITED STATES and the past 25 years in FRANCE.
- . Take representative samples of the final product in the container.
- . Conduct precise analysis that is compatible with the imposed criteria.

3.1 Specifications (see figure 4)

The American Society for Testing and Material (ASTM) has conducted a remarkable study in updating the specifications for natural UF6 (now called Commercial Natural Uranium - CNU). This new specification (C787) has been officially incorporated in the enrichment contracts of the US DOE and COGEMA.

C787 allows for a very slight contamination of natural uranium by uranium from reprocessing, shown by a maximum allowance of 20 ppm of U236.

This specification protects the enricher and the customers against accidental contamination that could occur during the conversion process or by heels containing U236.

The future ASTM C996 specification will apply to enriched UF6 from CNU. It will be more thorough than the current specification, chemically and isotopically. It will protect utility customers and their fuel fabrication customers by incorporating the industrial experience acquired so far.

In figure 4, we show the values that are recognized as acceptable by all French nuclear power operators.

3.2 Sampling :

Sampling should be a guarantee of the quality in the container. In addition to sampling during conditioning, UMF conducts :

- . Very precise controls of the state of empty containers, or containers with heels that could modify the quality of the material.
- . Sampling in the gaseous phase to analyze the isotopes, and in a liquid phase to isolate impurities of materials entering the COGEMA Pierrelatte site.

Depending on the customer requirements, the UMF can conduct sampling on all models of existing sampling bottles, including models 1S, 2S and P10 tubes.

Following customer demands, samples performed by UMF are sent to customer selected later for analysis.

The bottles are checked for tightness and cleanliness.

Maintenance operations include :

- . rough vacuum ($\sim 10^{-2}$ torr),
- . passivation with ClF₃,
- . nitrogen filling.

At the end of the five years, the valves are dismantled, the bottles undergo a chemical cleaning treatment, pressure and tightness test, and control of width measurements and welding.

3.3 Analysis :

The Quality Assurance Group manages the control and maintenance program and establishes rules.

These are described in detail in a QA program book. They are similar in many cases to the ASTM method as applied to up-to-date equipment.

Pack sampling bottle undergoes a subsampling according to the diagram shown in *Figure 5*.

Halocarbon and volatile fluoride content :

Processes :

- . Infrared absorption spectrophotometry (P.E.580)
- . Detection threshold : 2 to 10 Vpm, depending on molecules
- . Accuracy $\pm 5\%$ relative beyond detection threshold

Isotopic Analysis :

Processes :

A/ UF6 mass spectrometry : SMP 250

- . Simple or double 235U-234U-U236 standard
- . Detection threshold : 3 ppm
- . Repeatability (external) : 0.005 % to 0.05 %
- . Precision : 0.1 % relative concerning U5 or
- . Accuracy : 1 % relative for U4.

The accuracy only depends on the standards used. COGEMA owns a very wide range of standards. These standards were compared with those prepared by NBS, BNFL, EURATOM.

The compatibility of all these is better than 10^{-3} for U5.

B/ Mass spectrometry with thermo-ion source :

- . Analysis of minor isotopes : U4, U6
- . Accuracy : 2.10^{-3} (0.2 % absolute)

C/ Spectrometry :

- . U232 content (counting)
- . Detection threshold : 0.01 ppb
- . Repeatability : $\pm 5\%$ at 0.1 ppb

U assay :

Process :

- . Gravimetry
- . U₃O₈ weighing compared with an ultra-pure U metal standard submitted to the same operations.
- . Repeatability : 0.01 %
- . Test 4 times a year : conducted by a French authority (CETAMA) related with several European laboratories.

Impurities :

Processes : very various, according to elements to be counted, most particularly used are :

- . ICP spectrometry
- . Molecule absorption spectrometry
- . Atom absorption spectrometry
- . Accuracy and repeatability can only be specified one element at a time. It varies from a few ppb to a few ppm.

COGEMA is preparing for the future by developing a one-of-a-kind method which will replace traditional subsampling as well as the waiting time to obtain results on isotopic analysis.

4 - CONTAINERS

The quality of the UF6 product and the goal to fill up containers in a secure environment have led COGEMA to study these techniques and the cleaning methods. It is the micro fluorination method.

4.1 The transfer/sampling Unit of UMF is based on the "hot spot/cold spot" process on each production line.

One of the lines is equipped with a vacuum pump entirely dry and leaktight, with an important flow and manufactured by NORMETEX. Temperature and pressure operating conditions make this unit a particularly well-adapted device for evacuating possibly over-filled cylinders.

4.2 International and national regulations on pressurized equipment require security control every five years.

In FRANCE, containers designed following French specifications are under the control of the Ministry of Industry and Mine whose Mining Department is responsible for the program's implementation. The ASME regulates containers that are built according to foreign specifications. These two groups operate in the same way, following international regulations.

Therefore, the UMF has at its disposal a maintenance facility for 30B and 48Y containers with an annual capacity of 2,000 containers (Figure 6). Activities at the facility include :

- To perform internal cleaning through a highly-efficient process of high-pressure liquid unique aspersion : when necessary, the material contained in heels may be recovered. Cleanliness is checked by an authority not belonging to COGEMA.
- To carry out external maintenance of packings, through shot-blasting, painting and ultra-sonic checking of shell thickness, as well as systematic replacing of related valves and plugs.
- To perform hydraulic pressure tests (28 bar pressure), leaktightness test (7 bar pressure), vacuuming, nitrogen filling as well as final weighing and tare-weighing operations.
- To conduct specific checks such as dye penetrant testing, X-ray test, coating thickness test.

5 - SECURITY AT UMF

In FRANCE, nuclear safety and security are guaranteed by two safety laws applicable to radiation ionization and nuclear materials control : these two laws are 76.663 of July 1976 and 80.572 of July 1980.

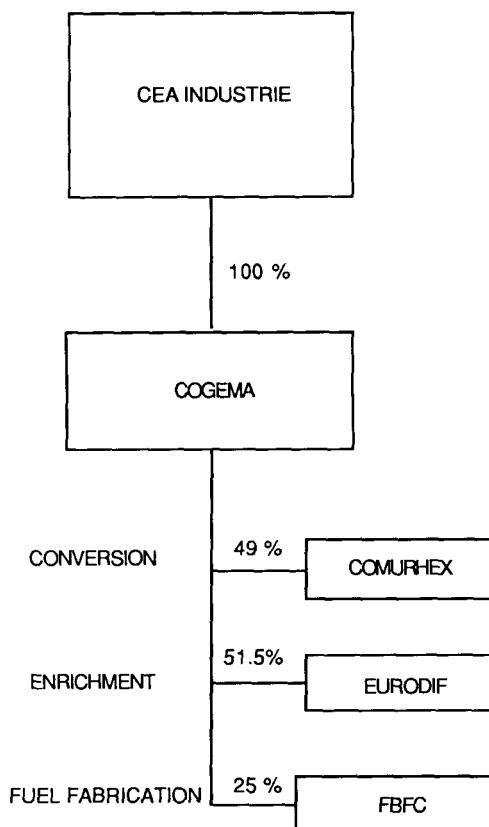


Figure 1 : Companies located on Tricastin site

These laws are applied through a series of decrees related to :

- Facilities and discharges to the environment
- Basic Nuclear plants
- Public safeguard
- Workers safeguard
- Capacitations
- Measures
- Nuclear material accountancy
- Physical protection of these materials
- Transports.

The Ministry of Industry is responsible to ensure the implementation of these laws with the technical support of the "Institut de Protection et de Sécurité Nucléaire" (IPSN). This group, which operates independently from the nuclear operators, conducts strict controls in liaison with the AIEA and EURATOM.

UMF is also organized along these lines : its storage areas are geographically separated and controlled continually by automatic surveillance systems. In addition, UMF benefits from the integration of the Tricastin site, which regroups all of the activities we have talked about today.

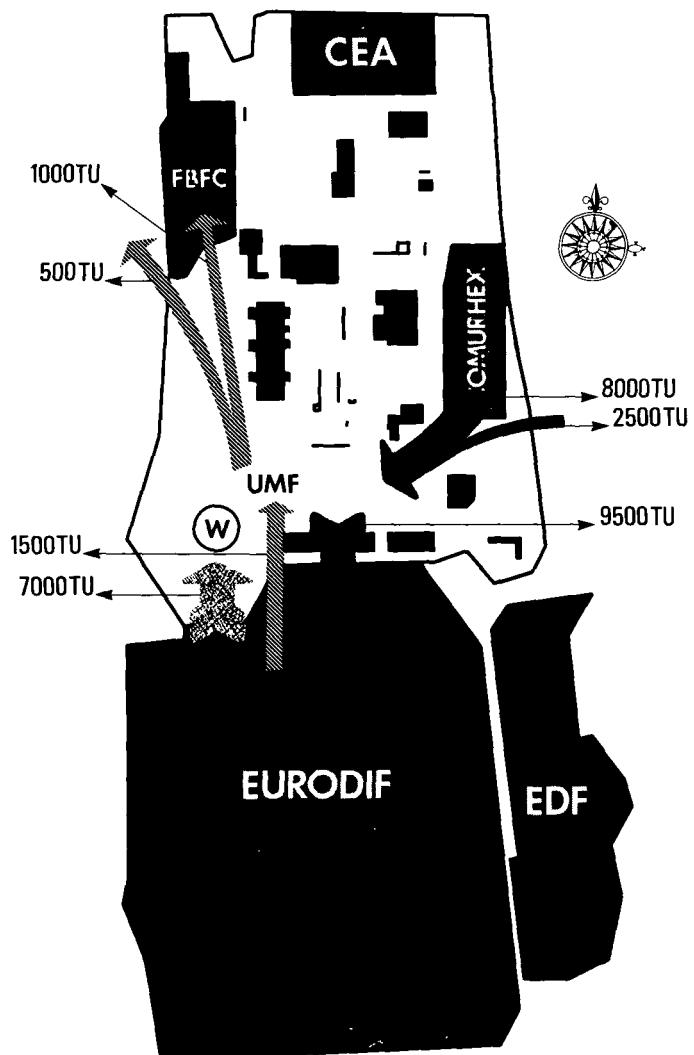


Figure 2 : Main UF₆ Flows in the Tricastin site

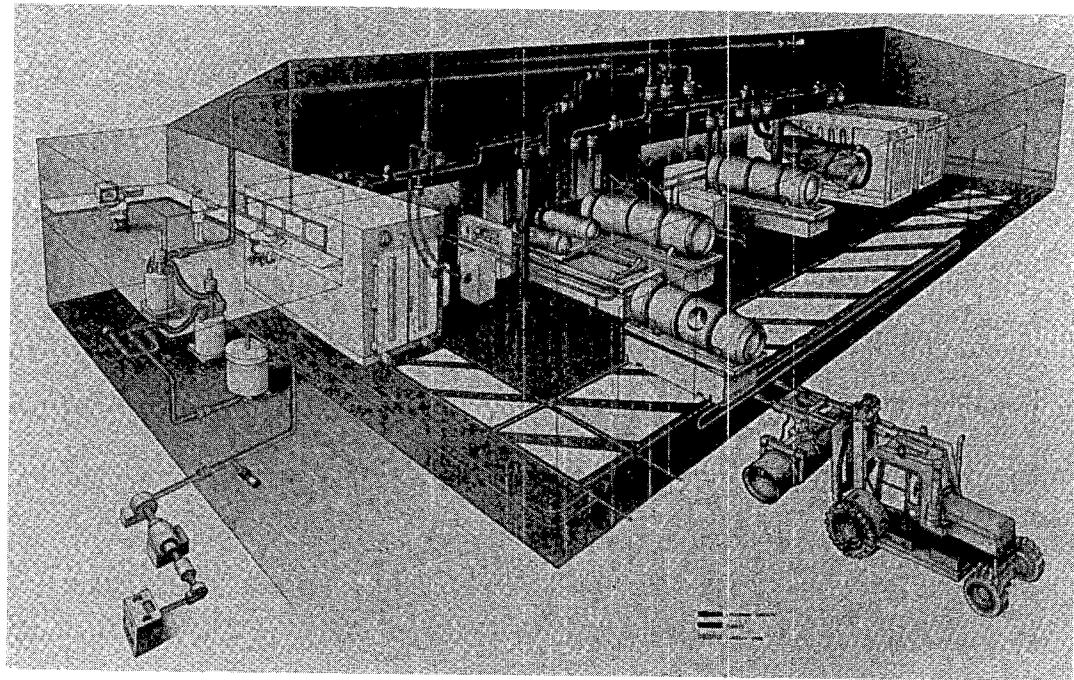


Figure 3

	ISOTOPY			CHEMISTRY		
	U232 ppb/ Total U	% U235	ppm/U U234	ppm/U U236	Mol %/UF6 Hydro Chloro Carbon	ppm/U Boron
"Virgin" Natural	0	0.711	55	0		
"Commercial" Natural	0,02	0.707 0715	< 65	< 20	< 0.01	< 1
Enriched Commercial	0,2	3 *	< 300	< 300	< 0.01	< 4

* As an example

Figure 4

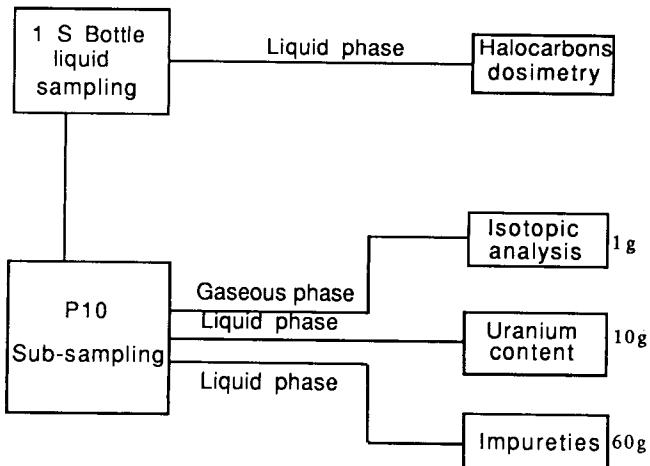


Figure 5

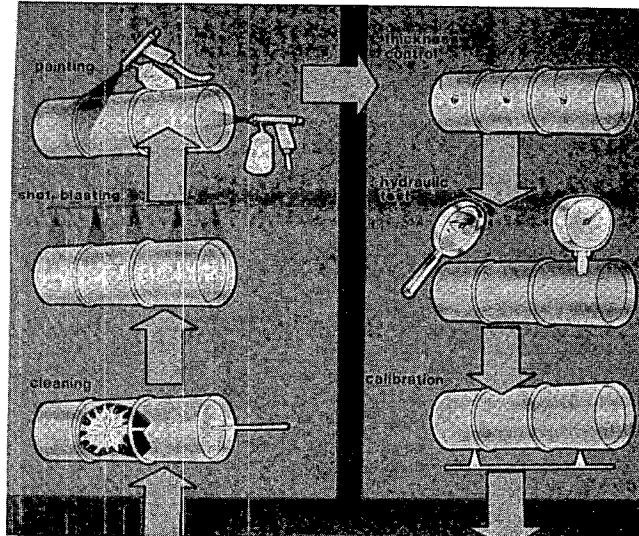


Figure 6

HISTORY OF UF₆ HANDLING COMMITTEE

W. E. Sykes
Paducah Gaseous Diffusion Plant
Paducah, Kentucky 42001

ABSTRACT

UF₆ handling operations at the Department of Energy (DOE) gaseous diffusion sites have been performed in a very safe manner since start-up of uranium enrichment plants. Department of Energy Oak Ridge Operations office (DOE-ORO) and gaseous diffusion plant contractor management were aware of the need to continue this safe level of handling performance and identify new approaches for further improvements of the overall safety of UF₆ handling. One such new approach was the formation of the UF₆ Cylinder Handling Committee in 1973. The first major effort completed by this committee was a comprehensive review of all UF₆ handling activities at the three gaseous diffusion plants. This review resulted in the development of 133 recommendations that describe procedures or operating methods for safe UF₆ handling operations. These recommendations are still applicable and are intended to serve as a basis for safe UF₆ handling practices at the DOE-ORO sites. Several of these recommendations have resulted in significant improvements in safety at the DOE sites and also to private companies who have adopted the practices into their operating procedures and are covered in this document. The UF₆ Handling Committee continues to be our main approach to interplant communication addressing problems and standardization of UF₆ handling related activities.

INTRODUCTION

From an overall standpoint, UF₆ handling operations at the DOE gaseous diffusion sites have been performed in a very safe manner since start-up of uranium enrichment plants. Thousands of UF₆ cylinders have been filled, emptied, moved, sampled, and transferred.

Over the years, there has always been a strong emphasis toward promoting safe UF₆ handling. Both DOE-ORO and gaseous diffusion plant contractor

management, Union Carbide and Goodyear Atomic, were aware of the need to continue this safe level of handling performance and hopefully identify new approaches to making further improvements or enhancements of the overall safety of UF₆ handling at the gaseous diffusion sites. In 1973 a new approach was identified as an idea that might result in such an improvement. This idea was implemented and what resulted is felt to have had a significant positive impact toward enhancing the overall safety performance of UF₆ handling at the gaseous diffusion plants. This new approach was the formation of a UF₆ Cylinder Handling Committee to be composed of a representative from each of the three gaseous diffusion plants. Each representative was both a knowledgeable expert in the various facets of UF₆ handling and a high-level management member.

The initial committee charter included the following points: review and evaluate UF₆ handling procedures and practices at the three sites to minimize the potential for UF₆ releases, review UF₆ release emergency procedures, recommend standard methods and procedures for UF₆ handling at all three sites, and recommend improvements in equipment and facilities to achieve maximum protection against UF₆ releases. The commitment to the high level of effort in this area was made because plant management and DOE realized proper and safe handling of UF₆ was critical to the emerging social consciousness of the uranium enrichment industry. During the late 1980s the title of this group was changed to the UF₆ Handling Committee. Even though the membership has changed over the years, the group's name has changed and the committee size has increased; it continues to serve as a coordinating group and communication vehicle on matters that relate to the original purpose of the group.

The first major effort completed by this committee was a comprehensive review of all UF₆ handling activities at the three gaseous diffusion plants to further ensure safe operations. This review, which required approximately 1-1/2 years to complete, resulted in the development of 133 recommendations that describe procedures or operating methods for safe UF₆ handling operations. These recommendations were published after receiving agreement by top management from the three enrichment plants toward implementation where applicable at each site. The implementation

Prepared by the Paducah Gaseous Diffusion Plant, Paducah, Kentucky 42001, operated by MARTIN MARIETTA ENERGY SYSTEMS, INCORPORATED, for the U.S. DEPARTMENT OF ENERGY, under Contract No. DE-AC05-84OR21400.

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period for these recommendations was approximately five years. Since that time the recommendations have received periodic status reviews at each plant to ensure actions that were intended to be taken were kept intact. The last status review of the 133 recommendations was made in 1986 at all the gaseous diffusion sites. These recommendations are still applicable and are intended to serve as a basis for safe UF₆ handling practices at the DOE-ORO sites. These recommendations and several other items developed by this committee since 1973 were incorporated into the DOE document ORO-651, Revision 4, "Uranium Hexafluoride: Handling Procedures and Container Criteria," in 1977. This information has provided guidance to groups worldwide.

The approach used in making the comprehensive review was to divide all UF₆ handling operations into 14 different areas for study. The following is a list of the areas reviewed and a summary of the number of recommendations that resulted:

AREA	NUMBER OF RECOMMENDATIONS
On-Site UF ₆ Cylinder Handling	16
UF ₆ Sampling	11
UF ₆ Feeding	11
Communications	6
Emergency Personnel and Equipment	8
Securing the Release	9
Decontamination	1
UF ₆ Transfer and Assay Blending	10
UF ₆ Withdrawal	11
UF ₆ Cylinder Integrity	15
UF ₆ Cylinder Valve Rebuilding and Testing	10
UF ₆ Cylinder Valve Replacement	5
Containment	7
UF ₆ Cylinder Pigtail Design and Testing	<u>13</u>
TOTAL	133

A subcommittee made up of representatives from the three plants for an area was assigned the task to complete a review and to identify important or key procedural steps or methods that if followed would prevent an incident that could potentially result in a UF₆ release. Past UF₆ release-related incidents at the gaseous diffusion plants were reviewed. Lessons learned and/or actions taken to prevent the reoccurrence of a particular incident were also used in the development of the recommendations. Each subcommittee provided their findings to the UF₆ Cylinder Handling Committee for review and approval. Once a recommendation was accepted and approved, it was understood that each plant would take prompt action where applicable to incorporate it in their way of performing a particular task or activity. A few of the recommendations that were included were already being performed at at least one of the sites but not consistently at all sites.

As previously stated these recommendations were written as a means to document an agreed-upon, approved, and consistent approach to the way various UF₆ handling related activities were to be performed. To review all of these one by one would take too long in a paper such as this. What I would like to do is cover several of the areas that are included in the famous 133. Several of these key methods or approaches to operations have resulted in significant improvements in safety at the DOE sites and also to private companies who have adopted the practices into their operating procedures.

The following are several examples of recommendations that are included in eight of the fourteen areas:

On-Site UF₆ Cylinder Handling

- Any cylinder found to be in an unacceptable condition, due to physical damage, or overfilled should be appropriately tagged immediately as this fact is determined. Tags used to warn or provide instruction are to be of a weather resistant type and secured with durable wire.
- Liquid cylinders are to be cooled a standard time of three days for 30-inch and five days for 48-inch before movement between buildings. Written instructions giving approval are required if it is deemed necessary to move a liquid cylinder before this time period has elapsed.
- A cold pressure check should be made on every cylinder before it is heated.

UF₆ Sampling

- UF₆ cylinders should always be heated with the valve open to some pressure monitor with high and low standards which will alarm in the event of an abnormal problem and shut off heat. Safety protection devices and alarms associated with cylinder heating should be tested and results documented.
- Cylinders should always be thoroughly inspected for damage prior to heating. Extra attention should be given to the valve and plugs. Inspection results should be documented.
- The hazard of using open flame torches to prevent valve plugging in sampling operations was recognized. It was recommended that alternate methods be developed to eliminate the use of torches.

UF₆ Feeding

- Cylinder valves should never be opened unless in full view of the person performing the operation.
- Respiratory protection is required during the opening and closing of a UF₆ system.
- Future designs for new autoclaves at the sites should include a device to manually close a cylinder valve inside a closed autoclave.

UF₆ Transfer and Assay Blending

- All plants should establish procedures to verify cylinder contents, assay, gross tare, and net weights prior to heating for transfer.
- Emergency pigtail crimping tools and/or emergency shut-off valves should be available for use as a means for emergency cutoff of a defective system.
- Pigtail design criteria should be established for materials of construction, methods of fabrication, and testing requirements.

UF₆ Withdrawal

- UF₆ detection devices should be installed in all withdrawal facilities where UF₆ is present at above atmospheric pressure. These devices should alarm locally and in continually staffed areas.
- In withdrawal positions where the cylinder is moved to the manifold on a movable cart, the drive mechanism on the cart should be interlocked such that the cart cannot be moved when withdrawal operations are under way. This interlock system must be automatic and not be dependent upon operator's action.

UF₆ Cylinder Valve Replacement

- Procedures for valve replacement should specify that valve installation comply with ORO-651 or ANSI N14.1.
- Hazardous or Safety Work Permits should be issued by personnel initiating the valve change. The condition of the cylinders, etc., contents, the safety equipment to be worn during job, and emergency equipment requirements should be specified on permit. A permit shall be issued for each cylinder.

Containment

- Remotely-operated or automatic-closure valves should be provided in withdrawal operations to terminate flow of liquid UF₆ in the event of a release.
- When new feed vaporization systems are required, provisions will be made for containment inside high-pressure autoclaves.

UF₆ Cylinder Pigtail Design and Testing

- UF₆ cylinder pigtails should be designed by Engineering personnel at the request of operating groups. The design should specify material of construction, fitting design, tubing size, design pressure, and temperatures and testing procedures. There should be as few fittings as possible in any given pigtail and made from one continuous length of tubing.
- Pigtails are to be field tested and inspected before each and every operation. The field test should include assuring the pigtail is a certified pigtail and there is no physical damage to tubing fitting or threads. Connector fittings should be inspected for plugging prior

to each use. Replace gaskets after each use. Gaskets fabricated from Teflon sheets are not to be used. Gaskets are to be of proper thickness and two gaskets should never be used in a fitting. The leak integrity of a pigtail should be checked by pressure and vacuum leak rates. The pressure test should be at a minimum pressure of 40 psig and not to exceed two times the operating pressure of the pigtail.

In the last several years the committee has met on several occasions to study major UF₆ release-related incidents that have occurred in UF₆ handling operations. Investigation reports were reviewed and/or oral presentations were made by personnel closely involved with the particular incident. Lessons learned and corrective action steps from each incident were used at each site where similar operations are performed to complete a review to insure that a similar incident would not occur in the future. It is interesting to note that all incidents that have occurred in recent years were primarily a result of violations of at least one of the original 133 recommendations. The committee has also addressed special problems or concerns that include UF₆ cylinder valve quality and the prevention of hydrocarbon oil in UF₆ cylinders. Input has been provided by the group to the ANSI 14 committees and to personnel involved with development of safety analysis reports, operational safety requirements, and many other engineering studies.

Approximately three years ago, a need was recognized to update ORO-651, Revision 4, and include additional information relating to UF₆ handling. Update of this document was done under the auspices of the handling committee. Prior to final printing of Revision 5, committee members reviewed and provided their approval. Revision 5 of ORO-651 was issued in September 1987. Additional information provided in Revision 5 includes the following:

- UF₆ Cylinder Pigtail Fabrication, Inspection, and Testing Flowsheet
- Definition and Handling Precautions of Overfilled Cylinders
- Cylinder Heel Reduction and Evacuation Procedure
- UF₆ Cylinder Valve Tamper Indicating Devices
- Supplemental Safe Fill Limit Guidance
- Expanded warning statement concerning the importance of cleanliness of cylinder and hazard of the reactivity of UF₆ with a hydrocarbon oil.

In 1985, a representative from Westinghouse Materials Company of Ohio in Fernald, Ohio was added, bringing the number of the committee to four. Representatives from Savannah River and Oak Ridge's Y-12 plant were added in 1986, making this a six-site committee. These additions to the original committee size were done at the request of DOE-ORO to further promote safety in UF₆ handling at the additional sites.

In addition to the Handling Committee activities already described in this paper, there have been several longer term, special emphasis, or limited scope problem areas that have been given attention through the use of subcommittees to the main group. These subcommittees have been appointed to work for periods as short as one to two months and a few have functioned for several years.

In February 1976 it became apparent to the UF₆ Handling Committee that each plant used slightly different criteria for acceptable cylinder valves as well as the cylinder valve bonnet nut. A UF₆ Cylinder, Valve, and Pigtail Committee was formally established to generate a list that would be approved for safe use.

Prior to this time several less formal interplant committees met and contributed to the development of UF₆ cylinders and valves. When this group was reorganized in October 1978 a charter was put together that included the following points.

- Maintain current specifications for procurement of cylinders and valves.
- Revise, as required, specifications to improve safety or meet regulatory requirements. Handling Committee reviews all changes and approves significant changes.

- Perform tests or evaluations, as requested by the Handling Committee.

On subsequent occasions this subcommittee has been requested to evaluate the use of noncoded and repaired UF₆ cylinders at the three gaseous diffusion sites, inspection criteria for UF₆ cylinder valves to determine acceptability at each site, and other items. This subcommittee continues to be active and effectively functions to address concerns and problems that address primarily UF₆ cylinders and valves.

Other subcommittees have dealt with special areas that include Containment Autoclave Design and Testing Procedures, UF₆ Cylinder Valve Closer Design, and Sample and Transfer Autoclave Lubrication Study.

SUMMARY

Safe handling of UF₆ continues to be of utmost importance at the DOE-ORO sites. The UF₆ Handling Committee continues to be our main approach to interplant communication addressing problems and standardization of UF₆ handling related activities.

CONFIGURATION CONTROL OF SAFETY SYSTEMS

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ABSTRACT

The introduction of safety systems to the gaseous diffusion plants gave rise to the need for some method of assuring that the testing, identification, and changes to safety systems are reviewed and controlled by authorized personnel. This need has been satisfied through the implementation of a configuration control system. The configuration control system involves each major division of the plant and provides a record of the tests, maintenance activities and a historical record of these activities for each safety system.

DEFINITIONS

The best way to understand the use of configuration control is to define the terms which are used in its implementation.

Safety Systems - Safety systems are defined as equipment and/or hardware used to assure that the operation of a facility will not cause unacceptable risk to the safety and health of employees and the public.

Safety Analysis Report - A safety analysis report is a document which provides specific information on how safety systems and administrative controls provide an acceptable level of safety for the operation of a facility.

Administrative Controls - A procedural mechanism which requires action by a person rather than by equipment to perform a function similar to a safety system.

Operational Safety Requirement - Requirements which define how the safety systems are operated and maintained and how administrative controls are implemented.

Configuration Control - A system which assures that the testing, identification, maintenance, and changes to safety systems are reviewed and controlled by authorized personnel.

CONFIGURATION CONTROL IMPLEMENTATION

The implementation of the configuration control system begins during the conceptual design phase of a project. Plant Engineering defines the safety systems required to assure the safe operation of the process or facility. The identification of safety systems are done in concert with the facility.

The existing facilities were given a thorough review as a part of the Safety Analysis program which was initiated by the Department of Energy (DOE) during the early 1980 time frame. The existing facilities have been modified to reflect the Safety System concept which was brought about by the Safety Analyses.

Engineering drawings are reviewed both in-house and by DOE. The proposed safety systems are marked on these drawings through the use of an open "star" design. When the project is completed and the system is accepted, the safety systems are identified with a solid "star" which is easily read.

A second set of drawings are prepared following the acceptance by the operating division which identifies each of the safety system components. The drawing legend contains the stores number, component name, reference drawing number, and instrument setting if applicable.

Safety systems are purchased through the use of "data sheets" which are prepared by the Engineering Division. The data sheets are sent to Purchasing who establishes a safety systems stores number and marks the purchasing requisition with a special code to assure that special inspection is provided for each component upon receipt at Paducah Gaseous Diffusion Plant (PGDP). The special stores number is a unique number for a particular component and remains with that component throughout its usage and storage at the plant.

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SAFETY SYSTEM IDENTIFICATION

When a safety system component is purchased and enters PGDP, it is inspected by the Quality Evaluation group and tagged with a "Spare Safety System Component" tag. The component is then delivered to the stores area where it is placed in a special safety system stores area. This area is fenced and locked to prevent usage of components without authorization. In order to alert operating and maintenance personnel of the presence of a safety system component, the components are marked with yellow tags to make them more visible. These tags are placed on the components following testing and acceptance by the operating division. The identification number remains on the component at all times while the component is in the system. The identification number is unique to that particular component and is found on the safety system drawings.

SAFETY SYSTEM REPAIR

Repairs must be made to safety systems from time to time. The repair of safety system components is one of the times when safety systems are vulnerable to mistakes. The Configuration Control System has built in safeguards to lessen this danger.

First, when these systems must be repaired, tested, or changed, special permits called Safety System Work Permits are issued by the operations supervisor to the maintenance craft performing the work. This permit does not protect the individual performing the work from any hazards which might be involved. Protection for those individuals performing work on a system is provided through the use of hazardous or electrical work permits. The Safety System Work Permit alerts the maintenance personnel that they are working on a system containing safety systems and special handling is required.

The second line of defense against mistakes is the use of the Component Tagging System. If the component cannot be field repaired, a new component is purchased from safety system stores and installed in the system. The spare component tag must be attached to the spare component or it will not be accepted by the Maintenance Division for installation in the system. When the component is installed, the spare component tag is destroyed, and the original, yellow safety system tag is reinstalled on the new component.

A new spares component tag is placed on the part which was just removed from the system, and the old part is delivered to the proper maintenance shop for repair. When it is repaired, the tag is marked to indicate that inspection is required. Following acceptance by the Quality Evaluation group, the part is delivered to stores where it is placed in safety system stores for storage in the proper catalog numbered bin.

Following repair of a safety system, the Safety System Work Permit is released by the proper maintenance supervision, and the system is tested to insure that it operates as designed.

If the component which had failed could not be repaired, the store's inventory would reflect the

withdrawal of the component and a new item would be purchased.

CHANGES TO SAFETY SYSTEMS

Changes to safety systems are not permitted until the changes are reviewed and approved by Engineering. The request for changes must be in writing from the Operations Division manager to the Engineering Division manager. The change is reviewed by the proper Engineering discipline, and the reply is made in writing back to Operations by the Engineering Division manager.

If the change requires revision of the safety documentation, DOE must also approve the changes.

The change control requirement of the Configuration Control System insures the approval and documentation of safety system changes and provides a means of insuring that drawings are updated when the changes are complete.

SAFETY SYSTEM DATA BASE

A safety system data base is maintained to schedule the testing of safety systems and to provide a historical record of tests and maintenance activity.

The data base is maintained on the ADP computer system with scheduled testing printed monthly. The system at PGDP is similar to the one at Oak Ridge Gaseous Diffusion Plant (ORGDP).

Data input starts with the first-line supervisor who completes a safety system data sheet each time any safety system activity such as maintenance or testing is performed. This sheet is mailed to a central area and is entered on the computer. The data sheet has areas for testing only and maintenance for maintenance activity. The system allows recall of any of the data which has been stored. The data can be recalled for delinquency reports, calibration and testing, maintenance activity, or other desired information.

The data base is the workhorse of the testing system for safety systems.

SUMMARY

In summary, we have briefly discussed the Configuration Control System as it is used at PGDP. The system provides many checks to insure that safety systems are not changed without authorization from personnel who are knowledgeable and that they tested on an established schedule.

The methods employed are tagging safety systems, properly storing components with limited access to them, and issuing safety system permits which alert the maintenance personnel that they are working on a system containing safety systems. Computerized records are maintained to provide scheduled testing and maintenance of safety systems.

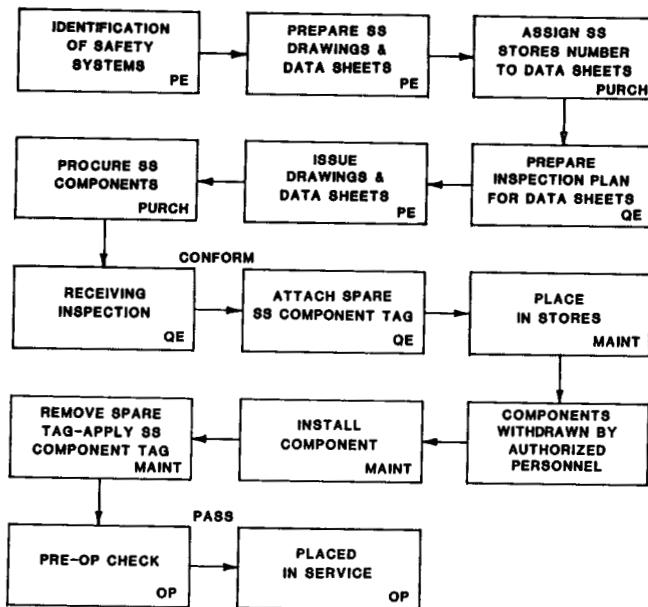


Figure 1. Normal Flow of Safety System

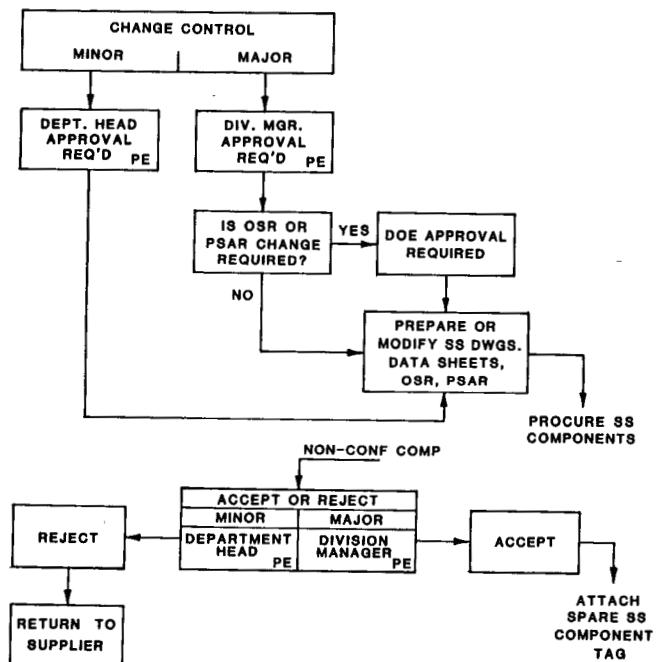


Figure 2. Failed Components of Safety System

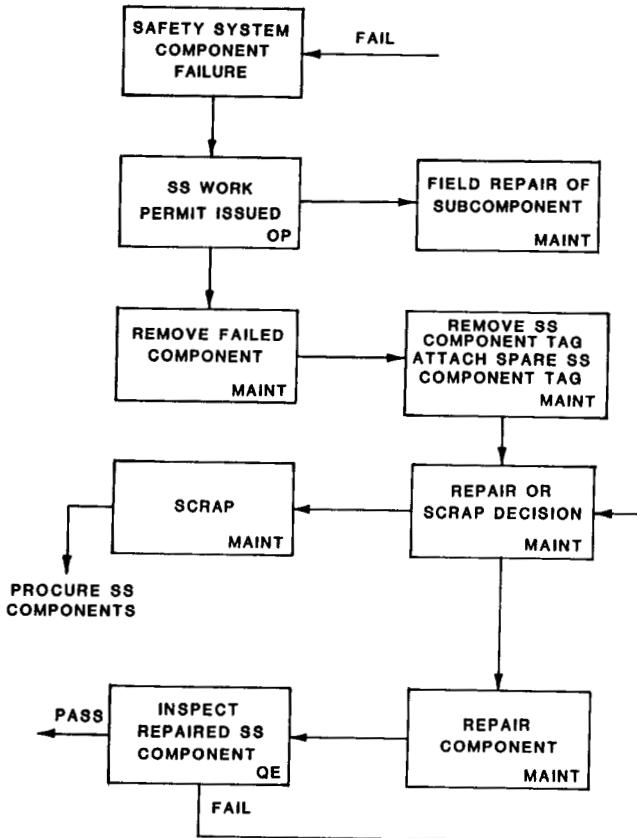


Figure 3. Changes to Safety System

URANIUM HEXAFLUORIDE - EMERGENCY PREPAREDNESS IMPROVEMENTS

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ABSTRACT

The United States Enrichment Enterprise has successfully handled uranium hexafluoride (UF_6) for almost 40 years. However, recent events and legislation have accelerated and emphasized the role of Emergency Preparedness. This paper will briefly describe several recent improvements in the planning/training, installation of new equipment, and public communications areas to further reduce the hazard potential of UF_6 . The improvements discussed are being implemented at the Department of Energy's (DOE) Gaseous Diffusion Plant, located at Paducah, Kentucky and operated by Martin Marietta Energy Systems, Inc.

The paper is organized into three major areas: Planning/Training, New Equipment, and Public Communication. The paper is designed as a collection of several emergency preparedness items. The brief review of these items should stimulate follow-up by interested parties.

Some of the planning/training items discussed are emergency squads and their training, the use of drills, tabletop exercises, emergency operations simulations, and full-scale field exercises utilizing mutual aid. Installation of new equipment will include the latest in impermeable suits and respiratory protection equipment, public warning systems, CO_2 systems, cylinder patch kits, meteorological stations, TV monitors, and other emergency operating center improvements. The public relations section will discuss the implications of installing a public warning system, involvement in local planning committees (SARA-III), and the use of a Joint Public Information Center (JPIC).

INTRODUCTION

"Emergency Preparedness Improvements" is a subject that encompasses more than can be mentioned in an

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article of this magnitude. The approach taken in this presentation was to collect and consolidate assorted improvements. This compilation should help put into perspective the risk of a UF_6 release and allow the reader to consider the value of emergency preparedness activities. The improvements will show both factors involved in risk assessment; likelihood and consequence, to be significantly reduced. The Paducah Plant is less likely, than in the past, to have a major UF_6 release and is better prepared should one ever occur.

PLANNING/TRAINING IMPROVEMENTS

The Paducah Plant has basically operated with the same emergency preparedness philosophy for over 35 years; this philosophy has been very successful. Line organizations have major responsibility within their areas. Prompt local action is required in these areas. Overall plantwide emergency direction is always provided by the Plant Shift Superintendent (PSS) who is the Plant Emergency Director. All shift organizations must be capable of handling emergencies at any time. Designated emergency response squads both local and plantwide must continually receive training and practice.

There have always been training and response drills conducted by all shift organizations. Currently, training has been expanded to include the following techniques:

- 40 hours of emergency training conducted by off-site experts annually.
- At least 24 hours of training by on-site personnel annually.
- Emergency response drills performed on all shifts.
- Emergency Management Exercises (EME) utilizing the emergency cadre, role players, emergency squads, and security forces conducted quarterly.
- Use of a new "tabletop" concept for exercising plans and procedures on-site, off-site, and in combination without the actual movement of emergency equipment.

- Major full-scale field exercises including off-site agencies. This years' exercise is titled "Readiness '88."

Planning concepts have also changed over the years. Currently planning has a much broader base. The use of consultants, the increase in mutual aid, involvement in local planning committees, and meeting the requirements of SARA-Title III. The area of Public Information is shaping emergency planning more than ever.

Conclusion of this section would not be complete without mention of Readiness '88, a complete test of the planning and training functions. Readiness '88 is a full-scale field exercise designed to test the coordinated emergency response of Martin Marietta Energy Systems, Inc., the U.S. Department of Energy (DOE), McCracken County, Ballard County, Illinois, and Kentucky and will be conducted May 14, 1988. The exercise is being coordinated by the Paducah Gaseous Diffusion Plant. The intent is to test and refine procedures designed to protect the public and employees in the unlikely event of a major plant emergency. Readiness '88 is a checkpoint along the way to an effective emergency preparedness program. Plans and procedures have been written or modified, tabletop exercises and response drills have been conducted. More than 300 people are expected to be involved in a test of joint capability. Readiness '88 is the most extensive exercise of its kind in the plant's history.

NEW EQUIPMENT

New equipment varies from windsocks to computerized meteorological plume analysis systems. In an attempt to organize, new equipment will be subdivided into three categories: prevention, detection, and mitigation. A listing and brief description is all that is possible for this article. Hopefully this brief review will stimulate follow-up by interested parties.

Prevention Type Equipment

- Cranes - anti-collision, radio-controlled, 360° rotation, and improved, double-hook lifting devices.
- Cylinder storage yards - increased storage areas allow "clear path" movement with cylinders at minimum height without moving cylinders over other cylinders. Yard design also restricts vehicular access.
- Containment systems - autoclaves, buildings, dykes, inflatable drain line plugs.
- Safety systems - Most critical systems, especially those operating above atmospheric pressure, have been modified to comply with the safety system concept. Items such as redundant water inventory, high pressure, low pressure, and a pressure and cylinder cart movement interlock are in place.
- Inventory reduction - HF storage tanks reduced from 28,000 pounds to 800 pounds.
- Atmospheric vents - solid sorbent trapping media added on all vents to the atmosphere plus

redundant instrument monitoring such as continuous wet chemical bubblers, real-time alpha particle analysis, infrared and acoustic analysis, and TV cameras.

- Normetex pumps - These internally sealed pumps have been installed in high pressure liquefaction applications, greatly reducing maintenance and the potential for leaks to the atmosphere.

Detection Type Equipment

- UF₆ sensors in process areas - monitor, alarm, and shutdown capability.
- Conductivity monitoring of autoclaves steam condensate - alarm and shutdown capability.
- Cylinder pressure - high and low alarm and shutdown capability.
- Autoclave pressure - high alarm and shutdown capability.
- TV cameras - visual surveillance and alarmed.
- Vibration detection - monitors, alarm, and shutdown capability of critical rotating equipment.

Mitigation Type Equipment

- New impermeable suit - Vautex, three-layer composite of viton, nylon, and neoprene.
- Respirators - full-face respirators, which utilize a larger cannister, rather than the one-half face are now being specified by the manufacturer. A new 10-minute escape only mask has been made available in specified areas.
- Large CO₂ system - 12,000 pounds of liquid CO₂ available to contain or control leaks from hot UF₆ cylinders.
- Prototype cylinder patch - much lighter in weight, more effective sealing design, and faster to install.
- Emergency operating center improvements - status boards, ring-down phones, personal computers, historian, telefax, back-up radio communications, improved maps and procedures, and improvements to the plant public address system.
- Meteorological stations - Computer-generated plume release models.
- Windsocks - 11 plant locations.
- Public warning sirens - two-mile notification of the public to shelter followed by Emergency Broadcast System (EBS) messages.

PUBLIC COMMUNICATIONS

Emergency preparedness improvements in the area of Public Communications are now mandated by Federal law. The Emergency Planning and Community Right-To-Know Act of 1986 (Title III) of the Superfund Amendments and Reauthorization Act (SARA) mandates that local citizens will

have ready access to information from chemical facilities. This information includes which chemicals are produced or handled and in what amounts, how much is released, and when spills have occurred. Title III also dictates that companies work with communities on Emergency Planning and give states, counties, and local fire departments information and a voice in chemical handling decisions. The Paducah Plant is building on years of experience in emergency preparedness and long established working relationships between the plant and off-site emergency agencies. Some of the highlights in public communication follow:

Public Warning System - A fixed siren system covering all of the two-mile immediate notification zone alerts residents to shelter and listen for further instructions on an EBS station. Prior to placing this system in service, a public information program which included new procedures for the plant and county emergency plans and door-to-door distribution of an information pamphlet to local residents was conducted.

Local Planning Committees - The plant Emergency Preparedness Coordinator serves as a committee member on the local planning committees of both neighboring counties. We have been declining the chairmanship of these committees, but are taking a very active and supportive role.

Joint Public Information Center (JPIC) - The Paducah Gaseous Diffusion Plant has recently established an off-site JPIC. The JPIC is a place at which all information concerning an emergency is officially released from all

involved agencies or groups. The JPIC is the centralized source of official information with the following responsibilities:

- Confirm emergency information with the media.
- Draft/issue press releases.
- Conduct press conferences.
- Staff rumor control phone banks.
- Manage live media coverage.

A well-equipped JPIC includes a large (100-200 persons) media briefing room, phones for the media, news hot lines, citizen hot lines, TV monitors, radio monitors, media work tables, TV camera areas, briefers' table, podium, technical experts, and agency public information officers' work rooms.

CONCLUSIONS

The United States Enrichment Enterprise has successfully handled uranium hexafluoride for almost 40 years. Improvements have continually been made which lessen both the likelihood and consequences of a major UF₆ release. We cannot be content with our past achievements and cease efforts toward further improvements. We must continue to balance the existing risks against increases in production costs. It behooves us all to continue to seek reasonable emergency preparedness improvements in a cost-effective manner.

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