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ASSESSMENT OF CONSOLIDATED UF_6 RELEASE STUDIES

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I. Executive Summary

The technical work addressed in this report had its origin in the technical support effort provided for the proposed packed-bed scrubber for the "Add-On" GDP at Portsmouth and a growing concern over the sites' ability to contain and predict the off-site impact of UF_6 releases. In the mid 1970s the technical managers agreed that scrubber studies would be conducted at Paducah and UF_6 knockdown work at Portsmouth. As a result of the SAR effort and dispersion modeling studies conducted by Battelle Laboratories, Columbus, Ohio, the need for data on the basic chemistry and physics of the interaction of UF_6 with atmospheric moisture was recognized. This assignment was directed to Oak Ridge in 1980. Prior to the consolidation effort of 1980, project action plans and objectives were developed with modest interaction between sites and without a directed set of objectives from management. Since that time, the Environmental, Health, and Safety (EH&S) Technical Program Management Team (TPMT) has insured against duplication of technical effort and provided project guidance on a limited scale. The authors recognize the wealth of information available in topically related technical reports. This report addresses the work in progress or initiated after the consolidation effort of 1980. The following project objectives (and their respective status) have evolved for the three sites.

A. PGDP

1. Objective: Characterize the performance of selected aqueous scrubbers in simultaneously removing UF_6 and its hydrolysis products from gaseous streams.

2. Status: A novel multichambered scrubber was found to remove both uranium and gaseous fluorides at greater than 99% efficiency under optimal conditions.

B. GAT

1. Objective: Determine the feasibility of various knockdown techniques for containing UF_6 releases in feed, withdrawal, transfer and sampling areas. Characterization of the reaction between UF_6 and atmospheric moisture and evaluation of the variables of UF_6 cloud formation and ultimate cloud fate have been consolidated to ORGDP.

2. Status: Scoping type studies have identified electrostatic precipitation as the most effective means of particle knock-down.

C. ORGDP

1. Objective: Determine (1) the physical and chemical characteristics of UF_6 hydrolysis products, (2) the mechanism of the hydrolysis reaction and the species and lifetime of the intermediates, (3) the morphology, change in morphology, and the rate of morphology change over a range of meteorological conditions.

2. Status: These objectives have been met over a number of release conditions where total UF_6 released, humidity, temperature of UF_6 at release, and release rate have been varied. The morphology of the UO_2F_2 formed has been characterized as a function of the conditions as has the change in morphology after release and the settling time of the cloud.

Major efforts have gone into the study of various types of UF_6 releases, their plausibility, probability, and consequences. These UF_6 release scenarios may be categorized as inside of structures or in the open. The release of UF_6 vapor in an autoclave (estimated at about 80 lb/min through a broken valve or pigtail) is considered a maximum credible occurrence. Scrubbers tested under these release conditions have proven capable of >99 percent removal efficiency for UF_6 and its hydrolysis products and improving visibility in the area to a level where emergency efforts can commence. The desired concentration of uranium and HF in the scrubber effluent and the degree of operational flexibility required by operating groups would constitute the primary requirements for which scrubber design would be chosen for low assay areas. If the decision is made to employ scrubbers for this application, additional scrubber studies could be conducted to minimize the quantity of scrubber solution generated in the event of such a release. With studies in progress, the chosen scrubber design could also be challenged by higher inlet uranium concentrations and very low humidity conditions. Any future scrubber work would require extensive renovation/upgrading of Paducah's test facility. Severe equipment restrictions would be needed to adapt scrubbers for use in high assay areas. Although not evaluated to the extent that scrubbers have been, electrostatic filters have been demonstrated to be feasible at Portsmouth. Additional testing would be needed to optimize operating parameters for containment applications.

The most serious, credible releases defined by SAR work are those involving liquid-filled cylinders. A major scenario for the release of liquid UF_6 inside a building (such as a ruptured cylinder in a feed facility) indicates that while the likelihood of occurrence is small, the potential impact to on-site and off-site personnel is great. Scrubbers are capable of significantly reducing the impact of this event.

For liquid releases outside a building, much can be learned from the release experienced at the French Comurhex Facility in July 1977. Upon breaking the valve (6 o'clock position) of a 7-ton cylinder at the threads (environmental conditions of 79°F, wind speed 20 mph), liquid UF_6 spilled to the ground, with subsequent formation of a lingering dense cloud, for 15 minutes before a wooden plug could be inserted to stop the leak. The use of water spray was attempted for release control but seemed detrimental to the effort. A blanket of carbon dioxide was effective in knocking down the cloud, probably due to limiting access of atmospheric moisture to the UF_6 and cooling effects, thus permitting access of personnel to the area. Comparing these results to the worst case scenario involving a 14-ton cylinder points to a significant concern over the control of an actual release and the time span involved. The dense cloud limiting visibility and continued release and reaction of UF_6 point to the need for an acceptable means of cloud knockdown and reaction control for spilled material. This same concern over the availability of efficient methods to deal with outside releases of liquid UF_6, was voiced as the first priority concern by Paducah Operations Division personnel when interviewed prior to preparation of this report. It is recommended that in addition to continued support of those release projects already budgeted and in progress at Portsmouth and Oak Ridge that funding and priority be provided to develop a set of "second generation" techniques to handle outside releases of liquid UF_6.

II. Response to Directed Questions

The following responses were prepared by the individual site members to the EH&S TPMT. These responses are directed toward the questions posed earlier by Donnelly.[1]

A. What were the original objectives?
B. Have the original objectives been met?
C. Are the original objectives appropriate in today's atmosphere?
D. Has the work significantly affected our ability to respond to a UF_6 release?
E. Has the work significantly increased our understanding of the mechanisms of UF_6 cloud behavior and knockdown?
F. What additional work is desirable?

The site project objectives and their status are presented in greater detail in the Appendix.

G. PGDP

1. The objective of the first phase of the scrubber effort at Paducah was to characterize the performance of selected aqueous scrubbers of the following types.

   a. Wetted Fiber Pad (WFP)
   b. Single-Stage Water Spray Chamber (SSW)
   c. Novel Multichambered Scrubber (MCS)
   d. Conventional Nozzleless Venturi Scrubber (NVS)

   The characterization involved in simultaneously removing UF_6, UO_2F_2, and HF from gaseous streams. A second phase of testing grew from the high removal efficiencies seen for the MCS system and the need to test a Conventional Nozzleless Venturi Scrubber (NVS). Second phase objectives included optimization of water consumption for the MCS, and evaluation of both systems (MCS and NVS) over a wide range of UF_6 concentrations including insufficient moisture for complete UF_6 hydrolysis.[2]

   A second supportive area of study centered on the identification of a suitable electrolyte and sampling technique for UO_2F_2 particle size analysis by Coulter Counter methods.

2. These objectives have been met with the scrubber studies completed in September 1981. The MCS system was found to remove both uranium and gaseous fluorides at greater than 99 percent efficiency at optimum conditions. Attempts to reduce water consumption below a weight ratio of 1.8 kilograms per kilogram incoming air resulted in lowered efficiencies especially at very dry ambient conditions. The NVS system provided removal efficiencies (96.4 to 99.7 percent) slightly lower than that of the MCS. The NVC system proved to be more sensitive to changes in release conditions in that there is less flexibility for making operational adjustments. Performance for the WFP was much lower for uranium and gaseous fluorides with removal efficiencies ranging from 69 to 98 percent. Application of the WFP system is thus marginal. Testing of the SSW system was terminated due to extremely poor performance as evidenced by penetration of the spray curtain and downstream deposition of UO_2F_2.

   Suitable electrolyte systems for UO_2F_2 particle size analysis were identified as LiI/Isopropanol and $LiBF_2$/Isopropanol. A major improvement in sample preparation was found to be collection of the sample on a filter medium followed by ultrasonic dispersion in the electrolyte.

3. The basic understanding of the capabilities of the various scrubber systems will stand as excellent core information. These studies provide confidence that off-the-shelf scrubbers remove particulate uranium at the same efficiency as they remove other particulate pollutants. Should employment of scrubber systems for GDP application be considered, a particular design can be chosen with confidence depending upon the required exhaust uranium concentration desired and the operational flexibility/sensitivity desired by the operating groups.

4. The work performed at Paducah, as defined by its objectives, was intended to enhance the capability of a GDP to respond to a UF_6 release through the use of scrubber systems. Since scrubber employment is not currently considered, the value of the scrubber effort lies purely in its potential application.

5. The objectives of the scrubber studies conducted at Paducah did not address these issues.

6. Additional scrubber studies can be justified if a decision is reached to employ scrubber systems for GDP application. The final testing considerations are (a) possible recycle of scrubbing solutions to minimize the volume of liquid subsequently requiring treatment (b) extension of the studies to higher levels of uranium concentration. The potential value of solution recycle rests in the reduction of waste solution treatment and solid waste disposal costs. These potential savings would be considered in light of the projected infrequent use of the scrubbers and the known costs required to renovate/modify the scrubber test facility and conduct the required tests. As part of these studies the proposed scrubber design should be challenged by higher incoming uranium concentrations to simulate more adverse release conditions.

H. GAT

1. In response to recommendations resulting from containment problems and the increased emphasis in containment for employee and public safety, knockdown studies were initiated at Portsmouth. The objective of this work was to determine the feasibility of various knockdown techniques for containing releases in feed, withdrawal, transfer, and sampling areas. Associated with this effort were the tasks of characterizing the reaction between UF_6 and atmospheric moisture and evaluating environmental variables of UF_6 cloud formation and ultimate fate. Due to delays in obtaining the environmental chamber and changes in personnel, this work was not started until 1978 and completed in 1981. Current studies include activities in two areas. One deals with UF_6 containment with the objective of developing techniques for mitigating, controlling, or eliminating existing or postulated credible accident hazards to plant personnel and/or public safety resulting from failure of plant components containing UF_6. The other deals with UF_6 release plume studies with the objective of providing assistance on plume tracking, analysis, and knockdown to the DOE/French information exchange team, primarily using the environmental chamber.

2. Within the scope of the original objective to determine the feasibility of various knockdown techniques, the objective has been met as electrostatic precipitation utilizing a charged stream of dry air were identified as the most effective. The associated task of characterizing the reaction between UF_6 and atmospheric moisture was consolidated to ORGDP and the task of evaluating environmental variables of UF_6 cloud formation and ultimate fate was partially completed at Portsmouth with the remainder of this effort being conducted at ORGDP. At Portsmouth, no association between settled UO_2F_2 cloud particle sizes and temperature or humidity effects in the range encountered in GDP operations were observed. The majority of particles analyzed were in the 0.5μ to 3μ range. In spite of the high density of UO_2F_2 particulate material, UF_6 hydrolysis clouds were observed to rise followed by diffusion. Turbulence was observed to shorten settling time by enhancing agglomeration; however, this would not be expected in open air environments where current UF_6 containment studies have dealt with evaluating commercial electrostatic air filters with reductions of 97 percent in airborne particulate concentrations being achieved. A rain simulation test has indicated little change in the airborne uranium content. Future activities in this project are to provide technical support and test environments for the development of fail-safe release alarm systems for cascade buildings and auxiliary facilities. UF_6 release plume studies are being conducted to provide cloud settling rate data and establish means of seeing UF_6 hydrolysis cloud growth, movement, and dissipation.

3. The original objectives are appropriate since information about various knockdown techniques is essential for a feasibility study dealing with improved containment in areas with higher potential for failure of UF_6 containing equipment. Objectives of current UF_6 containment and release plume studies reflect near-term concerns and are revised as needed to respond to changes in anticipated needs.

4. As defined by the objectives, the UF_6 containment work at Portsmouth was intended to evaluate the feasibility of various

knockdown techniques for containing releases. At this time, electrostatic concepts identified by this work are only candidates unique for future application in the GDPs.

5. Cloud behavior and the feasibility of various knockdown techniques, as defined by the objectives, are well understood for controlled environments. This information is applicable to small process areas where release clouds could be contained and knockdown techniques, as tested, utilized. Current UF_6 release plume studies are providing information which should enhance our understanding of cloud behavior in open field situations.

6. Additional electrostatic filtering studies could be justified if a decision to install knockdown facilities such as electrostatic filters for GDP applications is made. Final testing would be needed to establish optimum operating conditions, nuclear safety parameters, uranium recovery techniques, and other operational considerations.

I. ORGDP

1. The original objective was to formulate a model that would accurately describe a UF_6 release given the meteorological conditions, the amount of released UF_6, and affected terrain. This model incorporated the various heats of reaction, association, and phase change. To supply empirical information to meet the needs of the model and to better understand UF_6 releases, experimental work was performed at Portsmouth and ORGDP to determine (a) both the physical and chemical characteristics of hydrolysis products, (b) the mechanism of the hydrolysis reaction and species and lifetime of intermediates, (c) and the morphology, the change in morphology, and the rate of that change for a wide range of meteorological and total UF_6 released conditions.

2. These objectives have been met over a number of release conditions where total UF_6 released, humidity, temperature of UF_6 at release, and release rate were varied. The morphology of the UO_2F_2 formed has been characterized as a function of the

conditions, as has the change in morphology after release and the settling time of the cloud. Intermediates between UF_6 and UO_2F_2 have not been observed under normal release conditions and, if they exist, their lifetimes are considered to be very short. The HF formed appears to weakly adduct to the UO_2F_2, but is easily displaced by H_2O. The amount of hydration also is a variable yielding a large number of different UO_2F_2 x H_2O hydrates. A large effort was made to characterize the UO_2F_2 x H_2O formed, and to better understand the problems associated with its quantitative determination. The extent of hydration of HF and the onset of condensation as a function of HF and H_2O concentrations were determined.

3. The original objectives are appropriate since the basic information about cloud makeup is essential to the understanding of cloud behavior and fallout.

4. Outside of considering several remote sensing techniques to follow a release, particularly after dispersion has rendered it invisible; no response has been considered other than the currently in place actions to ensure the safety of plant personnel.

5. Cloud behavior in a controlled environment is well understood. Much less is known about cloud behavior in the actual environment. Cloud behavior is dependent upon the characteristics of the aerosol particles composing the cloud; thus, a better understanding of cloud behavior in the actual environment exists when wind and dilution are considered.

6. More experimentation is needed to evaluate the early stages of aerosol particle formation and the effects of environment on that formation. In addition, conditions approaching ambient need to be tested to better determine how our current results minimize an ambient release.

III. Summary

The project objectives for the UF_6 release work performed at Paducah, Portsmouth, and Oak Ridge are completed or currently in progress.

These studies have been coordinated by the EH&S TPMT since 1980 to prevent duplication of technical efforts. It is desirable to continue the plume cloud studies in progress at Portsmouth and the modeling studies under way at Oak Ridge. Further scrubber/electrostatic filtering studies are not warranted unless a decision favoring their employment is made. Additional scrubber studies would require extensive upgrading of Paducah's scrubber facility and an assessment of the resulting treatment/disposal requirements. Use of electrostatic filtering technology would require additional study to move beyond the feasibility stage and establish optimum operating conditions, uranium recovery techniques and other operational considerations. Scrubbers or electrostatic filtering would require evaluation of nuclear safety parameters.

Operations personnel were interviewed prior to preparation of this report to define their priority of needs concerning release technology studies. Technical support was requested in the following areas.

A. Advanced technologies/procedures for handling liquid release of UF_6.

B. Training and equipment to make use of the plume dispersion model and topographical overlays in determining the off-site impact of UF_6 releases.

C. Updating the existing UF_6 outleakage detection systems.

D. Determination of means for egress from facilities under release conditions where visibility is impaired.

It is felt that in these areas can the greatest strides be taken to reduce the risk to on-site and off-site personnel as well as the environment.

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APPENDIX: STATUS OF SITE OBJECTIVES

This Appendix has been provided in order that a more extensive listing by site of the individual projects and their objectives and status can be examined. An attempt was not made to judge either the statistical design of the studies or the quality of the work performed. Rather the data and conclusions provided by the authors are taken at face value.

TABLE 1 PGDP

Title/Document	Objectives	Status/Results
Evaluation of Aqueous Scrubbers for Removal of UF_6 Hydrolysis Products. (KY/L-713, Part 1)	Characterize the performance of aqueous scrubbers: 1. Wetted fiber pad (WFP) 2. Single-Stage water spray chamber (SSW) 3. Novel multichambered device (MCS) in simultaneously removing UF_6, UO_2F_2, and HF from gaseous streams.	1. Performance (removal efficiency) of the WFP varied from 69 to 98 percent and makes its application marginal. Inlet uranium concentrations for the trials ranged from 5000 mg U/m^3 to 16,000 mg U/m^3. 2. The SSW was the poorest performer. Studies were terminated due to penetration of the spray curtain and downstream deposition of UO_2F_2. 3. The MCS performs comparable (>99 percent removal at optimum conditions) for UF_6 systems as for other contaminants. a. Inlet uranium concentration for trials where injector gas was air was 7900 mg U/m^3 to 47,000 mg U/m^3. b. Inlet uranium concentration range for trials where injector gas was steam was 8500 mg U/m^3 to 38,000 mg U/m^3.
Advanced Aqueous Scrubber Studies (KY/L-1110, Part 9)	1. For the MCS: a. Test removal efficiencies over a wider range of inlet UF_6 concentrations including insufficient moisture for complete UF_6 hydrolysis. b. Optimize water consumption at atomizer chamber.	1. Over the relative humidity range of 15 to 75 percent, uranium removal was not found to be a function of relative humidity. a. Removal efficiencies remained above 99 percent for uranium and gaseous fluorides at optimum conditions. Concentrations for these tests ranged from 2660 mg U/m^3 to 61,400 mg U/m^3. b. Reduction of water consumption below a weight ratio of 1.8 kilograms per kilogram inlet air resulted in lowered efficiencies.

APPENDIX

TABLE 1 PADUCAH (CONTINUED)

Title/Document	Objectives	Status/Results
	c. Determine removal efficiency as a function of particle size.	c. Insufficient data generated. Based on manufacturers projections & efficiencies seen, a mean particle size of 3μ was projected with 10% wt less than 1μ strongly suspected.
	d. Investigate possible efficiency losses at low inlet UF_6 concentrations.	d. Over a wide range of ambient conditions and at low inlet UF_6 concentrations, removal efficiences remained greater than 86.6 percent.
	2. Evaluate a conventional nozzleless venturi scrubber (NVS).	2. The conventional nozzleless venturi scrubber provided removal efficiencies (96.4 to 99.7 percent) slightly lower than that of the MCS for uranium and gaseous fluorides. Selection for making operating adjustments (i.e. fine tuning) was not as great as for the MCS. These tests included uranium concentrations of 3,150 mg U/m^3 to 44,700 mg U/m^3.
UO_2F_2 Particle Size Analysis by Coulter Counter Method (KY/L-725)	1. Identification of a suitable electrolyte for UO_2F_2 particle size analysis by Coulter Counter Model TaII.	1. a. Suitable electrolytes were found to be LiI/Isopropanol and $LiBF_4$/Isopropanol. Both systems can be used with 50μ operature tube.
	2. Determine feasibility and workability of particle size analysis of UO_2F_2 in the 1μ to <1μ range by the Coulter Counter Model TaII using identified electrolyte.	b. Exclusion of water from the electrolyte system is essential. c. Solubility of UO_2F_2 in these electrolytes is low and only of concern for <1μ particles.
	3. Determine suitable sampling technique for collecting samples of UO_2F_2 particles from an air stream such that particle sizes are not altered and solubility problems are not encountered.	2. a. UO_2F_2 particle size ranged from 0.8μ to 40μ. b. Predominance of particles in the 0.8μ to 2.5μ range. c. Reduction of instrument noise can be achieved by the placement of shielding around the sample beaker.

1120102

APPENDIX

TABLE 1 PADUCAH (CONTINUED)

Title/Document	Objectives	Status/Results
		d. Additional study is needed to improve techniques for sealing sample beaker from humidity during analysis.
		3. Samples should be taken on filter media and then dispersed ultrasonically in electrolyte.

APPENDIX

TABLE 2 PORTSMOUTH

Title/Document	Objectives	Status/Results
UF_6 Containment Studies (GAT-T-3124, Part 6) (Mar. 1982)	1. Characterize the reaction between UF_6 and atmospheric moisture. 2. Evaluate environmental variables of UF_6 cloud formation and ultimate cloud fate. 3. Evaluate use of water, steam, CaO_2, Freon, air jet, boric acid, and air on UF_6 cloud knockdown and localization.	1. Effort consolidated to ORGDP 2. a. No association between UO_2F_2 cloud particle size distribution and temperature of humidity effects in range encountered in GDP operations. Majority of cloud particles in 0.3μ to 0.5μ range. Conclusions are for settled particles and many not apply to airborne particles. b. Turbulence in chambers may shorten settling time by enhancing agglomenation. Opposite may occur in open field with cloud distribution. c. In spite of higher Sensity of UO_2F_2· particulate matter, UF_6 release hydroloysis cloud immediately rises to ceiling of chamber followed by diffusion to fill chamber. 3. Reproducible UF_6 release cloud knockdown demonstrated (clear chamber in 5 minutes) with electrostatic-charged stream of dry air. Undisturbed cloud required 12 hours to 16 hours to settle.
Positive UF_6 Containment (520 Subdivision Project Status File) (Thru May 1983) and (GAT-T-3124, Parts 1,4, 5,6,7,8,10,&12) (FY-1982)	1. Develop techniques for mitigating, controlling, or eliminating existing or postulated credible accident hazards to plant personnel and/or public safety resulting from failure of plant components containing UF_6.	1. In progress - Feasibility of commercial electrostatic air filter demonstrated. Proposed use would be centralized location with ducting to remove contaminated air from anticipated release areas. a. Material balance indicated 40 to 60 percent of release recovery with 80 to 97 percent accounted for.

1120104

APPENDIX

TABLE 2 PORTSMOUTH (CONTINUED)

Title/Document	Objectives	Status/Results
		b. Indicated 96.8 percent reductions in measurable particulate UO_2F_2 in air returned to chamber. Rain tests indicate little change in airborne U content.
UF_6 Containment Studies UF_6 Release Plume Studies (520 Subdivision Project Status File) (Thru May 1983)	1. Provide assistance on plume tracking, analysis and knockdown to DOE information exchange team primarily using the environmental chamber. a. Determine cloud settling rates. b. Establish means of seeing UF_6 hydrolysis cloud growth, movement, and dissipation.	1. In progress a. Differential sampling point, vertical profile sampler and timed dropout samples collected. No significant concentration difference encountered through one foot differential sampler. Vertical profile sampler indicated gradual overall diminishing concentration at all levels. Timed dropout sampler indicated 0.17 in/min settling rate. b. Ultraviolet, thermographic, and optical methods, tried in chamber were unsuccessful. Ultrasonic reflection and possible forward looking infrared scanner will be tried to track cloud.

APPENDIX

TABLE 3 ORGDP

Title/Document	Objectives	Status/Results
Detemination of the Rate of HF Hydration and the Effects of HF on Moisture Condensation (K/PS-155)	Characterize solid products produced when UF_6 is released into air at specified relative humidities	1. UF_6 release experiments performed in small, static chambers at controlled atmospheric conditions; release chambers up to 8 cu. ft. in volume, UF_6 release of 5 mg to 230 mg RH of 2 to 100 percent.
Characterization of the Solid, Airborne Materials Created by the Interaction of UF_6 with Atmospheric Moisture in a Contained Volume (KP/S-144)		2. Solid products were collected and characterized by: electron microscopy, x-ray diffraction, laser light scattering, and cascade impactor using mass measurements.
Characterization of the Solid Product(s) Formed When UF_6 is Released into Ambient Air in a Contained Volume (K/ET-503)		3. Various morphologies and several compounds were observed dependent upon conditions of release. The particle size and degree of agglomeration were dependent on the relative humidity of the air and the temperature of the UF_6 at time of release.

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