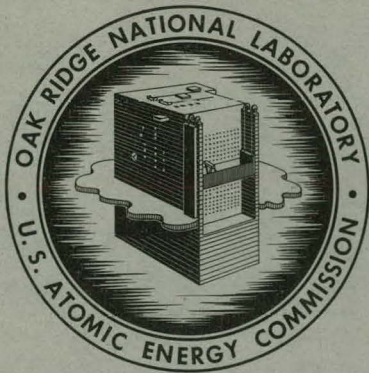


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MASTER

NUCLEAR SAFETY PROGRAM
SEMIANNUAL PROGRESS REPORT
FOR PERIOD ENDING JUNE 30, 1963



OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

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NUCLEAR SAFETY PROGRAM SEMIANNUAL PROGRESS REPORT
for Period Ending June 30, 1963

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Summary

PART I. PHYSICS

1. Reactivity Effects of Small Fuel Displacements in a Pool-Type Reactor

The "one-dimensional" critical experiment to facilitate comparison between diffusion-theory and transport-theory calculations of the reactivity effects of small fuel displacements in a pool-type reactor has been designed, and the experimental equipment has been assembled. The core slab will have transverse dimensions of 24 × 23.6 in. and will consist of about thirty 50-mil-thick elements spaced 200 mils apart. The slab will be contained in a 24-in.-wide, 40-in.-long, and 28-in.-high aluminum tank, in which water will be added as the moderator-reflector. Water-dump facilities will be included as a safety device.

2. Studies in Nuclear Safety of Fuel Outside of Reactors

Experiments were conducted to determine the source-neutron multiplication of EGCR fuel assemblies under conditions which might exist in transportation and storage. Twenty-eight fuel assemblies were arranged to yield maximum nuclear reactivity under conditions which might be expected for out-of-reactor environments. It was found that these 28 assemblies cannot be made critical when water moderated and reflected.

PART II. FISSION-PRODUCT RELEASE AND BEHAVIOR

3. Release of Fission Products on Out-of-Pile Melting of Reactor Fuels

Fission-Product Release from Aluminum-Uranium Alloys. — The release of iodine, tellurium, and

cesium from aluminum-uranium alloy heated in air increased with increasing burnup, temperature, and time at temperature. The release of rare gases was 98 to 100% complete when fuel specimens were heated in air for a few minutes at temperatures in the range of 700 to 1100°C. The release of ruthenium was low in this temperature range (700 to 1100°C) and seemed to be relatively unaffected by variations in time and air flow rate. Mixing steam with air had little effect on fission-product release. The release of cesium was greater in helium than in air, while that of tellurium at 1000 to 1100°C was very much less.

A highly radioactive mirrorlike deposit of material vaporized from highly irradiated aluminum-uranium alloy in helium or in a vacuum was found to be almost pure zinc which contained over 90% of the released iodine and nearly half of the released cesium. The proportion of cesium and iodine deposited from the helium stream in various parts of the furnace tube and downstream parts of the apparatus varied widely, indicating that cesium iodide compound formation was not an important factor governing the behavior of these fission products. Similar, but nonradioactive, deposits were obtained from unirradiated fuel samples, showing that zinc was a minor component of the fuel material. Particulate material caught on a filter when an aluminum-uranium alloy specimen was heated in helium at 1100°C contained zinc and aluminum. The particles varied in diameter from 0.04 to 4 μ . This temperature is above the boiling point of zinc, while the vapor pressure of aluminum is only about 10^{-3} mm, which indicates that some mechanism other than vaporization of pure aluminum must account for this major component of the fuel material becoming air borne.

Melting of UO₂ Fuel Pins by Centered Tungsten Resistors. — Unclad UO₂ fuel pins weighing 52 g each were arranged in a seven-pin cluster with the center pin irradiated at the tracer level and then

melted in purified helium to obtain fission-product-release data. In two experiments, with the seven-pin cluster, 28% of the center pin melted in one experiment and 90% in the other. Release of fission products from these arrangements was approximately one-half of that from similar melting tests previously reported where single unclad UO_2 fuel pins were melted. The deposit of fission products on the outer, unirradiated fuel pins did not account completely for the difference in release from the seven-pin and one-pin experiments. Release data corrected to 100% melting by direct proportion showed that 6 to 10% of the I, Te, and Cs were carried to the filter in the sampling system. Less than 0.03% of the Ru, Sr, Ba, Ce, and UO_2 were on the filter.

The third assembly which used the seven-pin cluster arrangement was constructed so that the helium sweep gas traversed a path providing longer and closer contact with the shields and base plate before leaving the furnace. The amount of I, Te, and Cs transported to the filter in this experiment was a factor of 10 less than with the other two cluster experiments, in which a direct gas path was available.

Effect of Time and Gas Velocity on Distribution of Fission Products from UO_2 Melted in a Tungsten Crucible in Helium. — The effect of time and gas velocity on the distributions of iodine, tellurium, cesium, and barium was studied. The total release was found to increase somewhat, under the conditions used, with increasing time for periods longer than 1.5 min. However, for a very short period of 0.4 min the releases were lower by factors of 2 or 3. The effect of doubling the gas velocity did not affect the transport significantly.

4. Release of Fission Products on In-Pile Melting of Reactor Fuels Under Transient Conditions

A review of the hazards analysis of proposed fission-product-release studies in the TREAT facility has been completed by TREAT management, and approval of the program has been given. The first two experimental assemblies, which are expected to be inserted in the reactor late in June, have been completed and tested. Stainless-steel-clad UO_2 fuel specimens will be pre-heated in the reactor to temperatures in the range 800 to 1200°C, and, after they are melted by fission heat in the reactor, the release aerosol will be sampled by admission to an evacuated autoclave.

5. Release of Fission Products on the In-Pile Melting or Burning of Reactor Fuels

In-pile experiments are being continued to study the release of fission products during simulated reactor accidents. Two types of experiments have been conducted in the ORR to simulate reactor accidents in which fuel elements were destroyed by melting or burning. One type consisted of melting or vaporizing a miniature stainless-steel-clad UO_2 fuel element in a helium atmosphere. In the other type, a miniature fuel element composed of spheroidal particles of uranium carbide coated with pyrolytic carbon and embedded in a graphite matrix was burned in air. In each case, fission and gamma heat raised the temperature of the fuel element high enough to cause destruction.

In the most recent UO_2 experiments the flow of helium sweep gas was increased from 400 to 800 cm^3/min ; otherwise, the conditions were the same as in the preceding UO_2 meltdowns. The results indicate no marked change with gas flow rate, and, in general, previous conclusions regarding release have been substantiated.

The distribution of fission products and uranium among the various regions in each experiment is being analyzed and interpreted in terms of the fractionation processes which govern the behavior of these materials. A study of fractionation should lead to a better understanding of both the mechanisms of release and the characteristics of released fission products. One result of the fractionation studies is the conclusion that in several of the experiments ruthenium follows the stainless steel cladding as the cladding melts into a puddle and subsequently vaporizes to other areas of the assembly. It is anticipated that this work should lead to the formulation of models which explain the observed distributions and which may provide a basis for prediction of the behavior, especially that leading to escape from a controlled system, of accident-released fission products under various conditions. A knowledge of fractionation mechanisms will aid in the recognition of the relative importance of the various factors which define a reactor accident.

In the experiments in which pyrolytic-carbon-coated uranium carbide graphite fuel was burned in air in the ORR, a large fraction of some of the fission products was retained inside the unburned fuel. The release of strontium, zirconium, barium, cerium, and uranium from the high-temperature zone of the furnace was very low, but large fractions of

iodine, tellurium, and cesium were released. Forty percent of the ruthenium was released from the furnace in one experiment and 5.7% in the other. These experiments indicate that the side of the fuel specimen with the least amount of uranium carbide burns preferentially.

6. Characterization and Control of Accident-Released Fission Products

In continuing the studies pertaining to the removal of ultrafine radioiodine-bearing particles from air streams using the diffusional deposition technique, evidence was observed that part of the airborne iodine was neither elemental nor particulate. The deposition and other data obtained indicated the presence in appreciable quantities of two distinct species of iodine other than elemental. One species appeared to be retained almost quantitatively at low flow rates of short duration by the inner surface of rubber tubing, and the other by activated carbon, but neither was quantitatively retained by silver metal. British workers in this field have previously reported that their data, observed for conditions not directly comparable, indicated some formation of iodine compounds when elemental iodine is released into atmospheric air.

Because of the dominant role of iodine in nuclear safety, elucidation of the origin and identity of each of the two species appeared to be a highly desirable objective, since such information is needed to interpret laboratory work and might even be needed to design adequate gas-cleaning systems for nuclear reactors. The diffusion coefficient of the compound reacting with rubber was determined to be approximately $0.05\text{-cm}^2/\text{sec}$, and mass spectrometry indicated the presence in the iodine source of two compounds having masses of 284 and 338 (amu), either of which values is not inconsistent with the diffusion coefficient of $0.05\text{ cm}^2/\text{sec}$. Thus, a fair amount of progress has been made toward establishing the nature of the compounds, and additional related work is currently under way, including investigation of alternate methods for elemental iodine preparation.

A method has been developed for characterizing radioactive aerosols in terms of their response to the processes of diffusion, interception, and inertial impaction by determining their distribution vs depth in fibrous filters under carefully controlled conditions. Quantitative theoretical analysis of the data corresponding to the diffusion regime has

yielded particle sizes which are consistent with electron microscopy, with photomicrographs, and with estimates based on the amount of activity in the aerosol and the number of particles measured by a condensation nuclei counter.

7. Fission-Product Transport Evaluations

Distribution of Fission Products Released from Molten Aluminum-Uranium Alloy. — The distribution of iodine, tellurium, cesium, and ruthenium as affected by atmospheric environment of air, helium, or 80% steam–20% air mixture during release was studied. Iodine released in air was apparently mostly in the molecular form, passed through the filters, and was absorbed efficiently by the charcoal traps. In helium, the bulk of the iodine was in another form associated with particulate material and was deposited on the filters and the furnace tube. The washout of vaporized iodine by the steam condensate appeared to be only 80% efficient in the mixture containing air. The total release of tellurium was low in helium, intermediate in steam-air, and high in air, particularly at temperatures above 1000°C . Below 1150° , cesium was found to be more volatile in helium than in oxidizing atmospheres. The release of ruthenium was sufficiently low in all cases as to prevent making definite comparisons.

Hot-Cell Containment Mockup Facility for Transport Evaluations. — A hot-cell facility has been designed, and its construction almost completed, for determining fission-product release from relatively large quantities of highly irradiated fuel materials. The gas-borne fission products released on heating fuel samples in different atmospheres will be transferred to a stainless steel tank that simulates a reactor containment vessel and held there long enough to permit study of agglomeration and plateout of fission products as a function of time and atmosphere, as well as the washout effect resulting from the presence of steam, foam, or other removal agents.

Retention of Radioiodine in a Mockup Containment Filter Unit. — An important feature of the Hot-Cell Containment Mockup Facility is the filter unit that simulates the “absolute”-filter-charcoal-bed arrangement proposed for use in certain reactor containment systems and also has provisions for drawing off portions of the gas flowing through the unit. The gas side streams, which will be removed before and after the “absolute” filter and also

after the charcoal bed, will be drawn through diffusion tubes to permit evaluation of the efficiency of the filter unit components in removing submicron particles and molecular vapors from the gas. Bench tests with this unit were performed to evaluate the iodine-removal efficiency of the types of filter material and charcoal proposed for use in the ORR containment system. The overall efficiency of the unit, which used two $\frac{3}{4}$ -in. beds of commercial-grade coconut charcoal in series, was found to be 99.99%. The diffusion-tube feature of the filter unit was not employed in these tests.

Identification of Chemical Species. — Experiments with the sealed-tube iodine equilibration apparatus gave values of 11.1 and 12.0 kcal for the heat of vaporization of iodine, with 0.20 mg and 5.1 mg total iodine, respectively, in the tubes. These studies have been discontinued because the heat-of-vaporization technique does not appear to be a promising method of characterizing chemical species in gas-borne accident-released fission products.

8. Iodine Removal from Oxidizing Gases at High Temperatures

Removal of Radioiodine from Air by Exchange with Inactive Potassium Iodide and by Platinized Aluminum Oxide. — Preliminary experiments were performed to test the iodine-retention capacity of noncombustible absorbents in air at high temperatures. These studies were prompted by the realization that charcoal beds, which are the chief deterrent to the escape of radioiodine from certain reactor-containment systems, may be destroyed or have their iodine-retention capacity impaired by exposure to hot oxidizing gases in a reactor accident. Inert potassium iodide and platinized alumina (hydroforming catalyst) were tested and were found to be quite effective for retaining iodine in flowing air at temperatures in excess of 500°C. Unplatinized alumina, Linde molecular sieve materials, and refrigeration-grade silica gel appeared to be less effective than platinized alumina in retaining iodine under similar conditions.

9. Attenuation of Fission-Product Escape by Intrinsic Processes

Certain intrinsic processes which tend to limit or attenuate the escape of fission products have not

been fully utilized in reducing the assumed consequences of a nuclear-reactor accident. Consequently, theoretical analyses of two such processes have been performed to determine if the attenuation effects are of sufficient magnitude to warrant further investigation or to warrant possible consideration in containment system design.

One of the processes analyzed was concerned with the decontamination resulting from diffusional deposition of radioparticulates in containment vessel leaks. The calculated decontamination factors were found to be extremely sensitive to both particle size and leak size; the higher decontamination factors are associated with the smaller particles and with the smaller leaks. Considering the single example of a containment vessel with a 1-in. wall and air escaping through leaks under a pressure drop of 7 psig, decontamination factors, defined as ratio of entering and exit concentrations, of 100 or higher were calculated for ≤ 0.3 - μ -diam particles when the nominal leak dimension was no greater than 5 μ . For larger particles or leaks, the results were much less favorable. The nominal dimension is the diameter for a cylindrical leak or the smallest dimension for a rectangular leak. The overall results obtained indicated that this effect is well worth careful attention.

The other theoretical study involved particle agglomeration and its effect on the penetration of filter systems in dual containment systems by fission products carried by particulate matter. A simplified model was analyzed in which the particulate matter was assumed to be pure iodine, and all particles with diameters less than 0.3 μ were assumed to penetrate the filters. It was shown that agglomeration would occur rapidly enough so that the particles, even though initially much smaller, would reach a 0.3- μ size in a short time (<20 min for typical water reactor systems). This time is directly proportional to the containment volume and inversely proportional to the power level. Assuming that the filter efficiency of the secondary-containment sweep-gas filtration system is zero for particles less than 0.3 μ in diameter, the fraction of the core activity released to the atmosphere during that time was calculated for some conditions of interest. For one reactor system treated, less than 10^{-5} of the iodine available would be released. It was shown that conditions existing during a reactor accident could not be as pessimistic as those of this model.

PART III. CONTAINMENT ENGINEERING

10. Nuclear Safety Pilot Plant

The Nuclear Safety Pilot Plant is an experimental facility to study the release, transport, and removal of fission products in a reactor containment vessel. Design of the facility was completed, and the installation work started in April is now about 60% complete. Design of remote handling tools, transfer casks, and other equipment for remote sampling and maintenance operations is virtually complete, and most of the equipment is being fabricated. Draft reports have been prepared on detailed operating procedure, experimental program, and safeguards.

11. Reactor Containment Handbook

A handbook for use in reactor containment systems is being prepared by ORNL and its subcontractor, Bechtel Corporation. Draft copies now exist on eight of the twelve chapters, and two of these have been distributed for external review. It is anticipated that the draft of all twelve chapters will have been completed and distributed by the end of this calendar year and that the handbook will be released next year.

PART IV. NUCLEAR SAFETY INFORMATION CENTER

12. Nuclear Safety Information Center

A Nuclear Safety Information Center for the collection, storage, evaluation, and dissemination of nuclear safety information is being established at ORNL. The Center's activities will initially be limited to the following six subject areas:

1. containment of nuclear facilities;
2. fission-product release, transport, and removal;
3. meteorological considerations;
4. nuclear instrumentation, control, and safety systems;
5. radioactive effluent control, monitoring, movement, and dosage;
6. reactor transients, kinetics, and stability.

Each of these areas is covered by a senior Laboratory scientist. The Center has assumed responsibility for a number of investigations in these areas.

PART V. RADIOCHEMICAL PLANTS

13. Radiochemical-Plant Safety Studies

Radioisotope source fabrication and Pu^{239} and U^{233} fuel fabrication in privately owned radiochemical plants are expected to increase greatly during the next decade. The potential economic loss from off-site damage due to accidents in such plants was evaluated. With the safeguards in design and operation normally included in a radiochemical plant, the worst contained accidents are not expected to result in a monetary loss due to damage to the surroundings which could exceed the limits of presently available insurance (\$60 million). As in the case of large power reactors, however, catastrophic-type accidents involving loss of containment in these facilities, although having such an extremely low probability of occurrence as to be considered incredible, could result in damage to the surroundings amounting to billions of dollars, particularly for the processing of normal inventories of Pu^{238} , Sr^{90} , and Pu^{239} . Uncontained accidents involving U^{233} or Kr^{85} , on the other hand, would result in potential liability of a million dollars or less.

The results of the study provide values of relative liability for plants that fabricate or process a variety of radioisotopes of current interest and provide rough estimates of the potential economic loss that could be incurred in major accidents. Radioactive material inventory, containment properties, meteorology, and surrounding population distribution all affect the potential monetary loss.

PART VI. SPACE SAFETY

14. Radiological Significance of Nuclear Rocket Debris

An investigation is under way on the potential radiobiological hazards to the general populace

arising from the operation of nuclear-powered spacecraft. Preliminary studies are concerned with the effects resulting from the contact of animal or human tissue with small intensely radioactive beta sources.

A computer program is being written to determine depth dose in any medium from spherical particles which contain mixed fission products.

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Introduction

This is the third of a series of semiannual reports describing the Nuclear Safety Research and Development Program which is being carried out at the Oak Ridge National Laboratory for the Research and Development Branch of the Reactor Development Division, USAEC.

This program has six objectives: first, establishing critically safe conditions for handling fissionable materials; second, obtaining data which are needed to assess realistically the consequences of accidents in reactors and chemical plants; third, developing and evaluating countermeasures to be employed in the event of accidents entailing radioactive materials; fourth, critically evaluating and compiling, in handbook form, information on reactor containment; fifth, collecting, interpreting, and reporting data in certain key areas of reactor safety; and sixth, studying health problems associated with nuclear rocket debris. To achieve these objectives, the following activities are being pursued.

1. The geometric changes which may occur in a plate-type fuel element in a nuclear excursion are being investigated to assess their importance from the standpoint of reactivity.

2. Conditions which are safe from a criticality standpoint for fabrication, storage, and handling of reactor fuels are being developed.

3. Out-of-pile and in-pile measurements are being made to assess the fractional releases of the fission-product inventories of important reactor fuels in the event of an accident. Those chemical and physical properties which bear on the removal of the released fission products from containment-vessel atmospheres and on leakage through a con-

tainment vessel to the environment are being studied.

4. Promising methods for removing fission-product aerosols from a containment-vessel atmosphere in the case of an accident are being investigated.

5. The small-scale investigation of fission-product release from fuels and of the behavior of the resulting aerosols will be substantiated on a larger scale in the Nuclear Safety Pilot Plant now being constructed. The pilot plant will be of sufficient size to better assess the time-space behavior of aerosols than is possible in the laboratory studies.

6. As an aid to the reactor designers, a Reactor Containment Handbook — a critically evaluated compilation of available information on this subject — is in preparation.

7. Accidents in fuel reprocessing and radioisotope production facilities may have more serious consequences than the accidents which are postulated for reactors. A radiochemical-plant safety study is therefore being carried out to improve the safety of these operations and to develop the information needed to assess accurately the consequences of accidents occurring in these facilities.

8. A Nuclear Safety Information Center is being set up.

9. In this report for the first time studies are reported on the evaluation of the safety problems associated with radioactive debris which results upon reentry of nuclear rockets.

Previous reports covering the period December 31, 1961, through December 31, 1962, are ORNL-3319 and ORNL-3401.

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Part I Physics

Neutron Physics Division

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1. Reactivity Effects of Small Fuel Displacements in a Pool-Type Reactor

E. G. Silver

As reported previously,¹⁻³ a program is under way to investigate the effect of small fuel displacements in plate-type reactors as a possibly significant mechanism of reactivity augmentation or compensation during transients large enough to induce substantial transient pressures. Initially, the experimental work was performed with the Bulk Shielding Reactor I, and the results were compared with diffusion-theory reactor code calculations. But it was found that there were no two- or three-dimensional codes available which could adequately handle the type of small-region, large-amplitude perturbations which were produced in the reactor by the selective removal of single fuel plates and the addition of fuel in the spaces between fuel plates. It was therefore decided to perform a series of simple-geometry ("one-dimensional") critical experiments which could be compared with one-dimensional calculations. Both transport-theory codes and diffusion-theory codes are being used so that their relative adequacies can be studied. The most recent phase of the

program has been to perform a number of the calculations, to establish the experimental parameters, and to assemble the experimental equipment.

The parameters for the experiments were established from the results of approximately 60 one-dimensional calculations by use of the MODRIC code, a one-dimensional multigroup diffusion code with Goertzel-Selengut hydrogen slowing down. It was found that with the available fuel plates (50-mil-thick unclad Al-U²³⁵ alloy plates containing 4.28×10^{-2} g of U²³⁵ per cm²) the transverse dimensions of the proposed core could not be very large without making the core thickness so small that the removal of a single fuel element would cause too great a reactivity change. Conversely, it was found that if very small transverse dimensions were adopted to increase the core thickness, then serious calculational difficulties would arise in computing the transverse neutron leakage. The code permits either the specification of a single transverse buckling, which is then the same for all energy groups and regions, or the buckling may be obtained from the code itself for each energy group and each region by specifying the dimensions of the core and calculating an extrapolation distance appropriate to each energy group and material by use of the diffusion coefficient calculated for that energy group and region. Critical core thicknesses determined by the two methods

¹E. G. Silver *et al.*, *Nuclear Safety Program Semiann. Progr. Rept. June 30, 1962*, ORNL-3319, pp 5-10.

²E. G. Silver *et al.*, *Neutron Phys. Div. Ann. Progr. Rept. Sept. 1, 1962*, ORNL-3360, pp 12-16.

³E. G. Silver, *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, pp 3-4.

differ in direct proportion to the number of elements explicitly specified in the code, and the magnitude of the effect increases markedly as the transverse buckling increases. Figure 1.1 illustrates the effect.

The transverse dimensions finally chosen for the fuel elements (and hence the core) are 24 by 23.6 in., which appear to be the most advantageous compromise between edge-leakage problems and critical core-thickness requirements. The effect of the spacing of the elements on the core thick-

ness was investigated in calculations for center-to-center spacings of 200, 250, 300, and 400 mils. Figure 1.2 shows the results. Although the smallest spacing yields the thinnest critical core slab, it also yields the core with the largest number of fuel elements, so that this element spacing will result in the smallest reactivity effect from the removal of a single plate. For this reason, a 200-mil spacing was chosen for the experiments; however, the aluminum tank which will hold the elements will permit some core loadings with

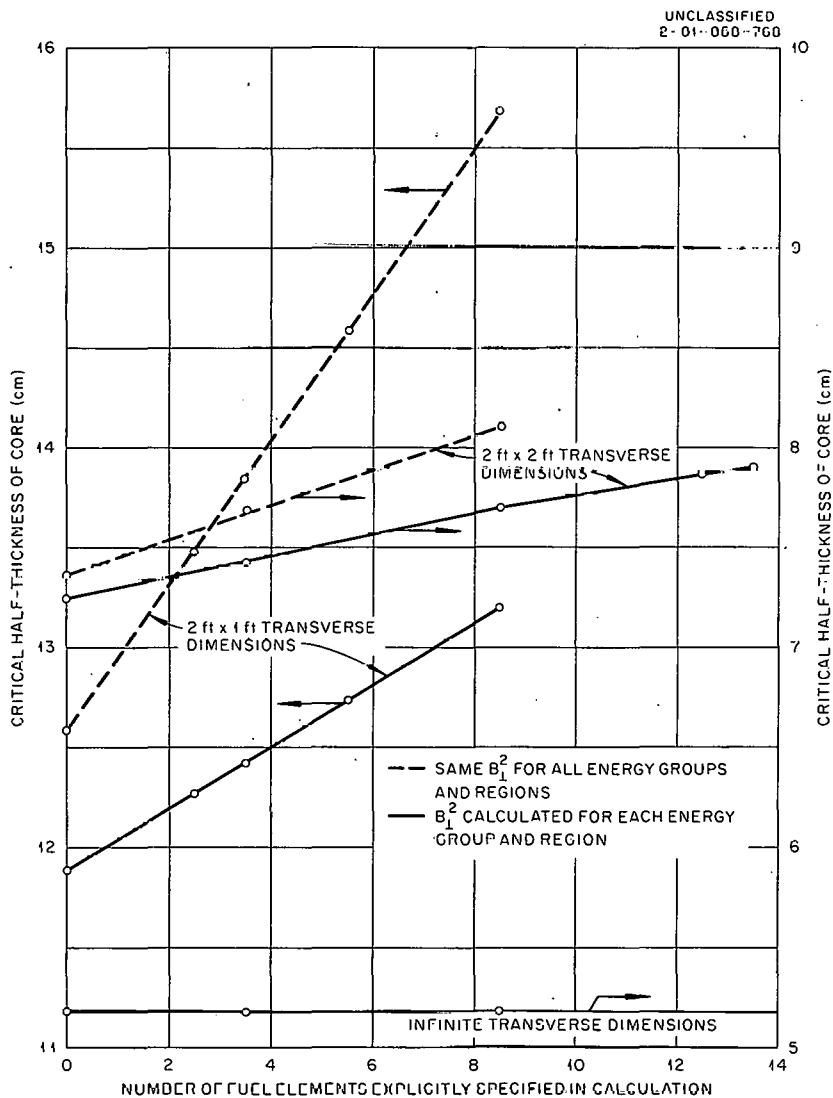


Fig. 1.1. Effect of Transverse Dimensions and Buckling Calculation Method on Critical Core Thickness (Calculated by MODRIC Code).

larger spacings also. This small element spacing, which produces an undermoderated core, will also maximize the diffusion vs transport effects and is hence advantageous for purposes of this investigation.

The experimental arrangement will be as shown in Fig. 1.3. The procedure will be a stepwise loading of fuel elements, each consisting of eight 2-ft-long plates, using water height as a reactivity shim, until a critical slab is attained with the water level just to the top of the fuel elements. The reactivity vs water-height curve for a small range of water heights from about 23 to 24.5 in. will be measured by inhour methods so as to serve as a small-reactivity adjustment device. If necessary, some of the eight plates in the outermost fuel elements will consist of dummy plates so as to bring the critical water height as nearly as possible to 23.6 in. (to the top of the fuel elements). In the reflector region at each end of the core stack, there will be 3-in.-diam drain pipes with flat valves connected to the safety circuits which

can rapidly dump the water into a dump tank. The water between the fuel plates will also flow out, though at a slower rate, through two 0.75×1.0 in. channels at each bottom edge of the tank. These channels are provided to assure hydrostatic equilibrium and thus equal water level between all fuel elements, since no other flow channels for the water between the plates are available. The fact that the core water level will probably drain rather slowly compared with the level drop rate of the reflector water does not represent a safety hazard, since this thin core slab (about 6 in. thick) is largely reflector-moderated in any case.

All the unperturbed-core diffusion calculations are complete, and the perturbed-core diffusion calculations, as well as the transport calculations for both perturbed and unperturbed cores, are under way. The number of transport calculations required will be substantially smaller than the large number of diffusion calculations, since all the parameter-study work leading to the design of the experiment has been completed using the diffusion code.

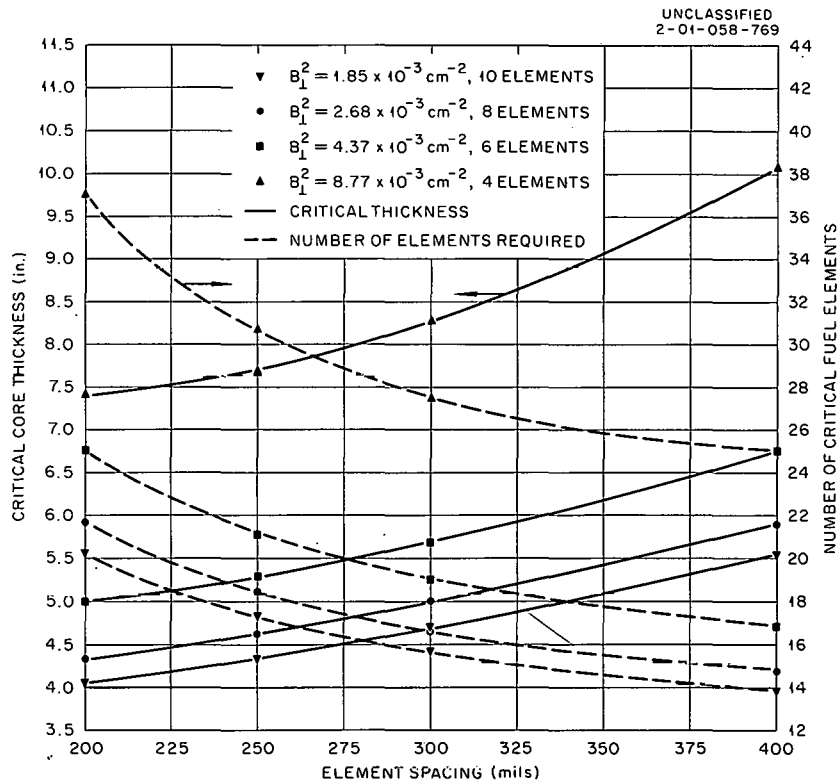


Fig. 1.2. Effect of Element Spacing on Critical Core Thickness and Critical Number of Fuel Elements.

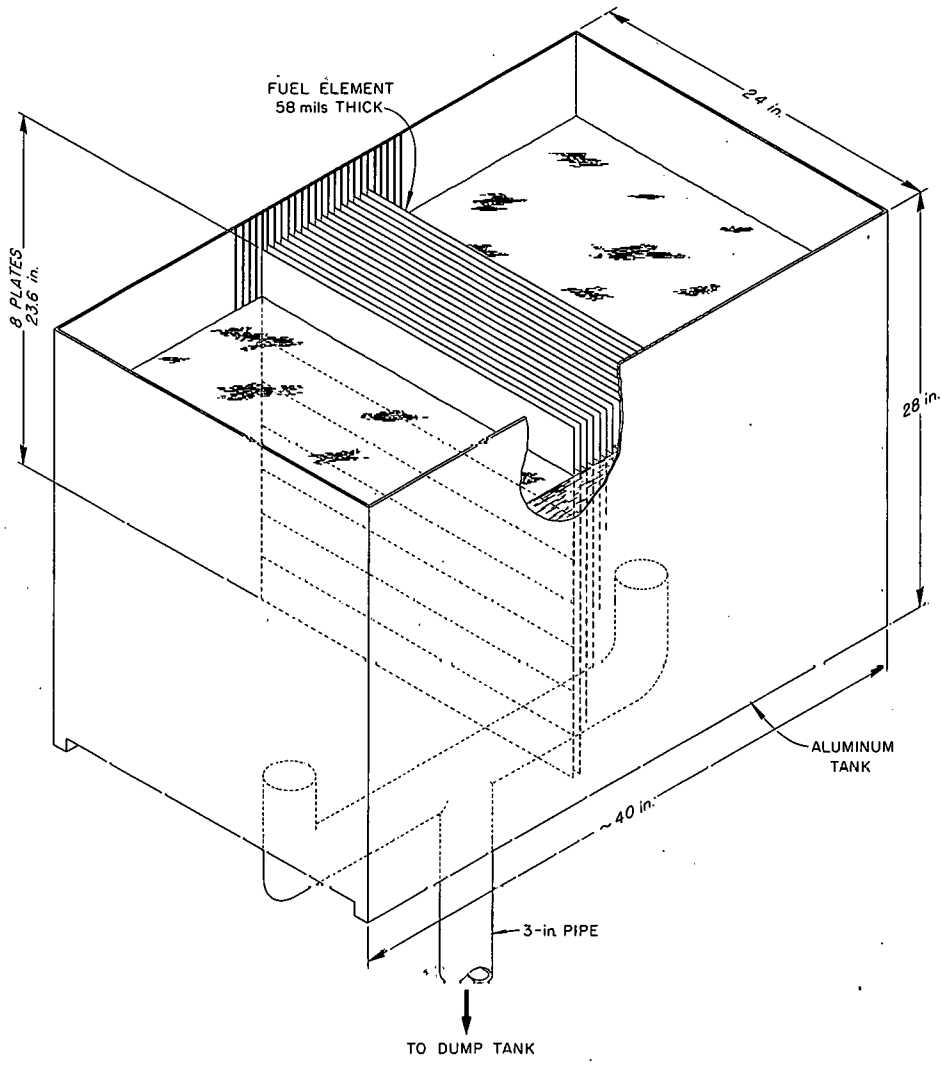


Fig. 1.3. Diagram of Arrangement for "One-Dimensional" Critical Experiment.

2. Studies in Nuclear Safety of Fuel Outside of Reactors

A. D. Callihan

In order to establish some measure of the critical number of the fuel assemblies for the Experimental Gas Cooled Reactor as a basis for the specification of their transport and storage, a series of experiments was performed in the Critical Experiments Facility. The conditions of the experiments were purposely made those which might be practically expected in an out-of-reactor environment which yields maximum reactivity. The assemblies were, therefore, submerged in water as they might accidentally be in transport or in storage. In one variation, water was excluded from within each assembly as it may be by packing materials of low hydrogen density.

The fuel, 2.46%-U²³⁵-enriched UO₂ hollow pellets, was sealed in type 304 stainless steel tubing (20-mil wall) about 0.7 in. in diameter and 27 in. long. A fuel assembly consisted of seven of these fueled tubes supported by stainless steel spiders within a 1-in.-thick, 29.4-in.-long graphite sleeve with an outside diameter of 5.0 in. The average U²³⁵ content of each of the 28 assemblies available for these experiments was 214 g. The fuel assembly has been described in detail by Samuels.¹

The assemblies were arranged in triangular lattices, as shown in Fig. 2.1, on a plastic table

¹G. Samuels, *Nucl. Sci. Eng.* 14, 37 (1962).



Fig. 2.1. Triangular Lattice Arrangement of Fuel Assemblies for the EGCR.

set on the floor of a tank which could be remotely filled with water and drained. A neutron source was placed between the fuel tubes in the central assembly.

It was experimentally determined that the arrays were most reactive with the assemblies in contact. Extrapolation of the inverse source neutron multiplication curve obtained with assemblies in con-

tact indicated that as few as 40 assemblies might be critical when moderated and reflected by water. However, this number is uncertain because of the long extrapolation of the multiplication produced by the available 28 assemblies. Excluding the water from within assemblies resulted in an observed neutron multiplication not significantly different from that with the assemblies filled with water.

Part II

Fission-Product Release and Behavior

Reactor Chemistry Division

W. R. Grimes, Division Director
G. M. Watson, Coordinator

3. Release of Fission Products on Out-of-Pile Melting of Reactor Fuels

G. W. Parker
G. E. Creek

W. J. Martin
R. A. Lorenz

FISSION-PRODUCT RELEASE FROM ALUMINUM-URANIUM ALLOYS

Effect of Temperature, Time, Atmosphere, and Burnup

Current interest in fission-product release from aluminum-uranium alloys was prompted by a review of hazards associated with research reactors which use plate-type fully enriched aluminum-uranium alloy fuel. Local concern is exemplified by the planned use of charcoal traps and absolute filters in the containment systems of the High Flux Isotope Reactor and the Oak Ridge Research Reactor. Since no release data on fuel materials of this type with a significant degree of burnup were available, it was considered necessary to conduct some release experiments to determine the effect of this variable, which has been shown to be very important in other types of fuels. An evaluation of other pertinent variables including atmosphere, time in the molten state, and temperature was conducted.

The release measurements were conducted mainly with a highly irradiated and long-cooled LITR-type plate that had been irradiated to 23.6% burnup of the U^{235} . Fuel specimens in the form of punched disks were heated in a quartz tube under the appropriate atmosphere for periods of either 2 min

or 1 hr (see Tables 3.1 to 3.3 and Figs. 3.1 to 3.4). Evaluation of the time variable was somewhat complicated by the experimental procedure, which required a total of about 10 min above the melting temperature for the heating and cooling cycles. The time interval reported included only the period at the maximum temperature. Since air and steam atmospheres are of principal concern to these reactors, most of the results were obtained in air.

The release values obtained were unexpectedly high, probably indicating the influence of burnup on release level. Release data at tracer-level irradiation were reported previously,¹ and recent data, obtained by exposing specimens in air at 805 and 910°C, are contained in Table 3.4. Comparisons of the results given in Tables 3.1 and 3.4 reveal that the releases from the specimen irradiated at tracer level are substantially lower than the corresponding releases from the specimens irradiated to 23.6% burnup of U^{235} . For example, the releases of iodine and of the rare gases are lower by approximately a factor of 3, and the releases of tellurium and cesium are lower by a factor of 4. The rate of release of the rare gases

¹G. E. Creek, W. J. Martin, and G. W. Parker, *Experiments on the Release of Fission Products from Molten Reactor Fuels*, ORNL-2616, p 11 (July 22, 1959).

Table 3.1. Release from Uranium-Aluminum Alloy Melted in Air (23.6% Burnup of U²³⁵)

Temp (°C)	Release (%)					
	Gross Gamma	Rare Gases	I	Te	Cs	Ru
After 2 min ^a						
700	2.3	97.9	37.8	0.3	3.1	0.02
800	3.1	99.4	78.6	0.2	3.8	<0.1
900	5.2	99.98	91.9	2.1	6.2	0.1
1000	6.7	99.83	97.3	<9.7	8.8	0.23
1090	12.0	99.99	98.4	44.8	12.4	0.6
1145	16.0	100.00	94.2	62.0	18.6	0.36
After 60 min ^a						
700	3.3	97.7	58.0	<0.14	3.5	<0.02
800	4.45	99.5	84.7	0.73	5.93	0.025
900	6.3	99.95	95.3	2.9	9.16	0.22
1000	18.1	99.98	92.8	16.6	23.3	0.1
1090	16.1	99.98	98.3	78.4	37.8	0.03
After 60 min with High Air Flow ^b						
840	5.3	~100	94.6	1.5	6.5	0.13
870	8.1	~100	95.8	4.0	6.9	0.7

^a250 cc/min air flow.

^b3000 cc/min air flow.

was found to be a function of burnup (Fig. 3.4). The profiles or histograms shown in Fig. 3.4 also indicate a multiple burst phenomenon with a dominant cooling burst at the tracer level and a large single initial heating burst for the high-burnup material. Evidently, at high burnup levels, the higher concentration of fission-product gas leads to efficient sweeping from connected pores, suggesting a spongy melt. This program will be essentially complete when tests with intermediate-burnup-level fuel materials (3 to 15%) are conducted; however, the effect of several variables on the transport of fission products will be studied in the Hot-Cell Containment Mockup.

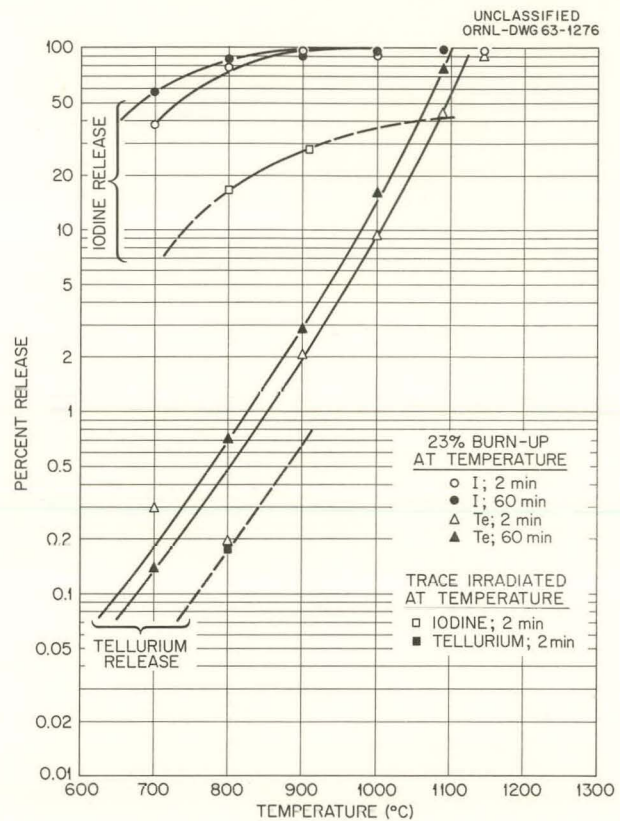


Fig. 3.1. Effect of Burnup, Time Molten, and Maximum Temperature on Tellurium and Iodine Release from Uranium-Aluminum Fuel Melted in Air.

Table 3.2. Release from Uranium-Aluminum Alloy Melted in Steam-Air Mixtures^a (23.6% Burnup of U²³⁵)

Temp (°C)	Release in 2 min (%)					
	Gross Gamma	Rare Gases	I	Te	Cs	Ru
700	0.92	98.3	27.0	<0.03	0.62	<0.02
800	2.5	99.5	76.8	0.26	1.1	0.1
900	6.8	99.9	90.6	5.65	6.5	0.45
1000	10.6	~100	95.6	22.6	11.0	0.5
1085	25.5	~100	96.8	67.9	30.5	0.8

^a250 cc/min air and 1000 cc/min saturated steam.

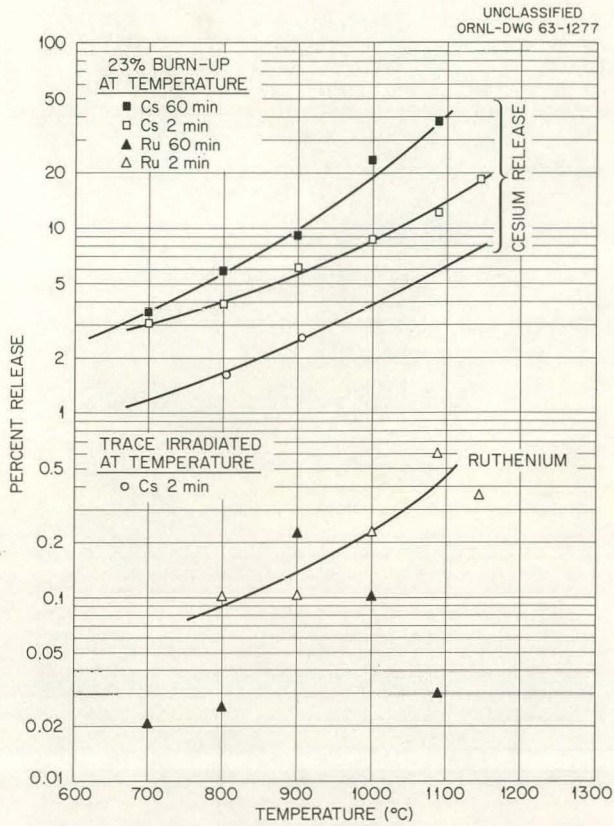


Fig. 3.2. Effect of Burnup, Time Molten, and Maximum Temperature on Ruthenium and Cesium Release from Uranium-Aluminum Fuel Melted in Air.

Table 3.3. Release from Uranium-Aluminum Alloy Melted in Pure Helium^a (23.6% Burnup of U²³⁵)

Temp (°C)	Release in 2 min (%)					
	Gross Gamma	Rare Gases	I	Te	Cs	Ru
800	7.4	99.5	29.8	5.3	13.0	0.18
900	13.5	~100	52.8	4.3	20.8	0.08
1000	23.0	~100	82.1	2.9	47.7	0.19
1105	40.7	~100	82.4	2.9	69.5	0.25

^aFlow rate, 250 cc/min.

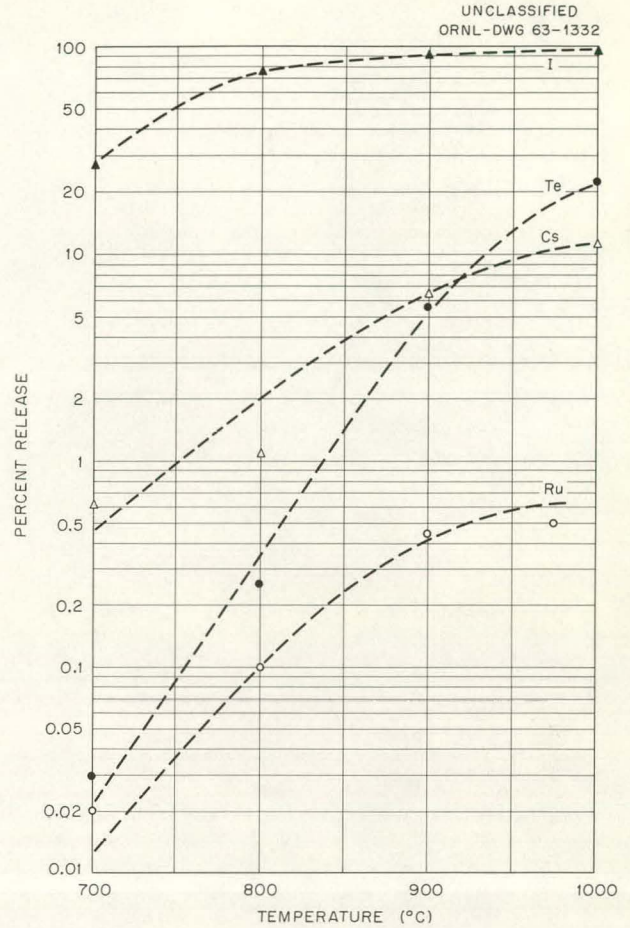


Fig. 3.3. Release of Iodine, Ruthenium, Tellurium, and Cesium from Aluminum-Uranium Fuels Melted in Steam-Air Mixtures; Effect of Temperature at Constant Burnup (23.6% U²³⁵).

Table 3.4. Release from Tracer-Level-Irradiated Uranium-Aluminum Alloy Melted in Air^a

Temp (°C)	Release in 2 min (%)					
	Gross Gamma	Rare Gas	I	Te	Cs	Ru
805	0.89	37.2	16.7	0.18	2.6	0.004
910	1.3	54.0	28.8	0.03	1.6	<0.0019

^aFlow rate, 250 cc/min.

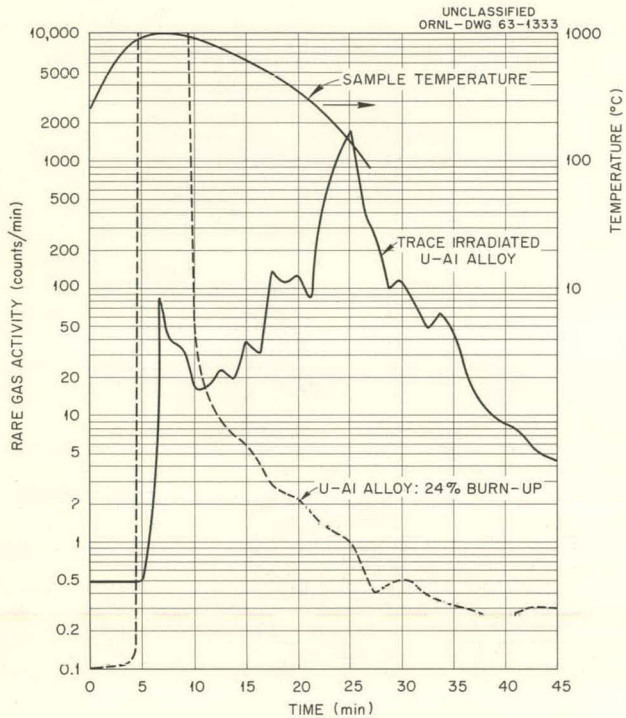


Fig. 3.4. Rate of Rare-Gas Release from Molten Uranium-Aluminum Fuel as a Function of Burnup.

Identification of a Material Vaporized from Aluminum-Uranium Alloys

When aluminum-alloy release experiments were conducted in pure helium a visible plateout of a highly radioactive mirror deposit was observed in a section of the quartz furnace tube just outside the furnace where the temperature varied from approximately 360 to 150°C. Since very little of the radioiodine passed through the absolute filters in these experiments, it was thought that the two phenomena might be associated.

As a means of obtaining a less-radioactive sample of the deposit, a sample of about 2 g of alloy containing no short-lived iodine activity was distilled and the mirror plate (Fig. 3.5) was isolated for redistillation, since cesium activity still remained a significant radiation source. After vacuum distillation, a somewhat decontaminated specimen was vaporized into a capillary for x-ray diffraction analysis by J. H. Burns, who reported that the material was metallic zinc. Spectroscopic and activation analyses confirmed this report. As a final test, a sample of clean, unirradiated alloy



Fig. 3.5. Metallic Distillate Vaporized from Highly Irradiated Uranium-Aluminum Alloy.

fuel plate was then heated in helium to see if the mirror occurred, since zinc was a likely impurity in aluminum. The unirradiated specimen produced an identical mirror which is shown in Fig. 3.6.

The distribution of fission products released from irradiated aluminum-uranium alloy specimens was determined as follows: material leached from the furnace tube up to the section containing the zinc deposit was the first sample; material from the short section of furnace tube containing the zinc was the second sample; the remainder of the furnace tube furnished the third sample; and washings

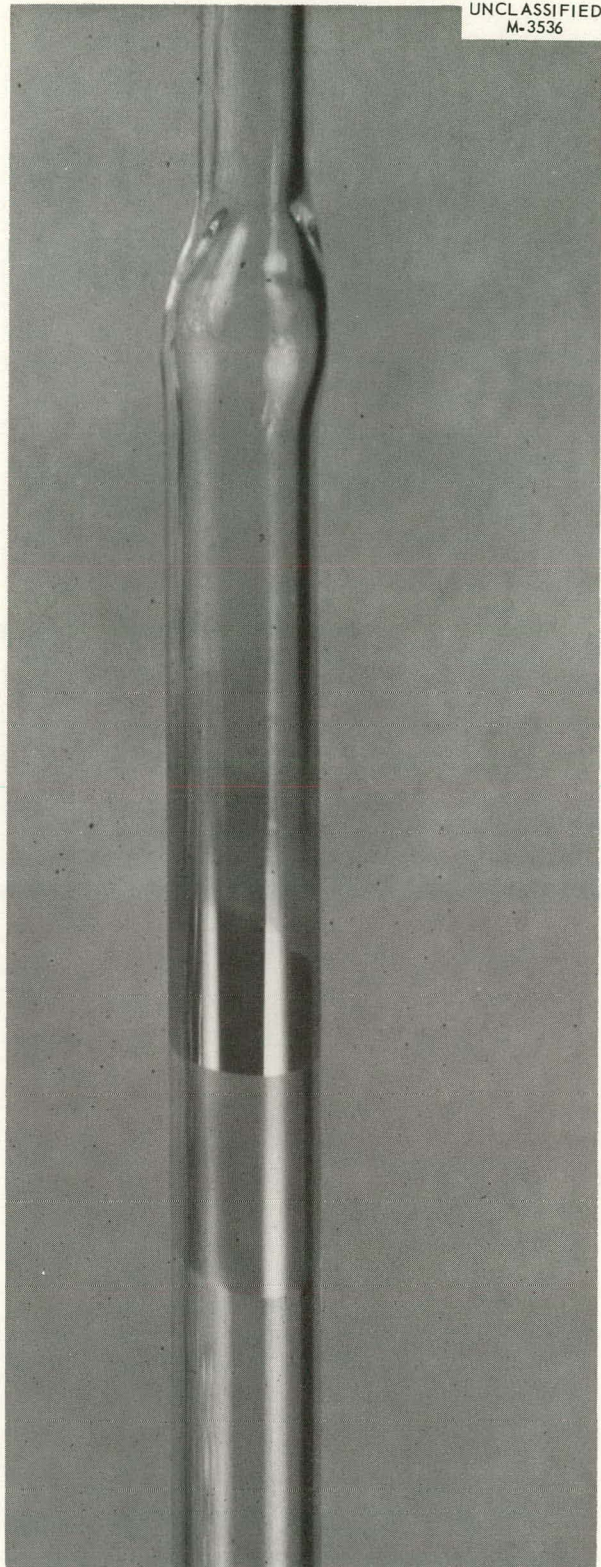


Fig. 3.6. Metallic Zinc Vaporized from Unirradiated Uranium-Aluminum Alloy.

from the tubing that connected the furnace tube to the Millipore filter housing was the fourth sample. The Millipore filter and the charcoal trap that followed the filter were also analyzed. The results given in Table 3.5 show that a large fraction of the iodine and nearly half the cesium were associated with the zinc deposit.

Since some investigators believe that iodine may be released from irradiated uranium heated in pure helium as a uranium iodide, the uranium content of the zinc deposit recovered in one experiment was determined and found to be 0.01%. The total iodine content of the deposit was not determined, but it was undoubtedly many times the amount required to combine with the trace amount of uranium present. The widely varying proportions of cesium and iodine found in the various parts of the apparatus, shown in Table 3.5, make it seem unlikely that a major part of these species was combined as CsI. However, the small amount of iodine that reached the charcoal trap indicates that iodine was in the form of a compound, possibly ZnI_2 .

In an experiment in which an aluminum-uranium alloy specimen was melted at about 1100°C in helium and about 17% of the iodine was caught on the absolute filter, presumably as particulate material, a dark deposit was observed on the filter. Electron diffraction of a portion of this filter indicated that the deposit contained both zinc and aluminum. The particles were found to vary in size from 0.04 to 4 μ .

Table 3.5. Distribution of Activity Released from Irradiated Aluminum-Uranium Alloy in Helium

Location of Activity	Percentage of Total Activity Released		
	Gross Gamma	Iodine	Cesium
1st sample	38.0	1.2	37.6
2d sample	45.3	92.7	43.6
3d sample	7.5	1.0	7.4
4th sample	0.46	0.68	0.45
Millipore filter	11.0	3.9	10.9
Charcoal trap	0.004	0.50	

MELTING OF UO_2 FUEL PINS BY CENTERED TUNGSTEN RESISTORS

G. W. Parker R. A. Lorenz

Realistic simulation of reactor loss-of-coolant accidents has been attempted on a small scale in out-of-pile experiments by melting UO_2 pellets with centered tungsten resistors. This experimental technique has progressed from the initial arrangement for melting a single fuel pin of three UO_2 pellets to an apparatus for melting a cluster of seven fuel pins consisting of four UO_2 pellets each.

When the center of a single fuel pin is heated to the melting point, the outer UO_2 surface usually does not melt, because of radiation to relatively cold surroundings. Fuel pins arranged in a cluster, as shown in Figs. 3.7 and 3.8, permit heat from the outer pins to radiate to the inner pin and cause nearly complete melting of the UO_2 in the center

pin. This type of experiment simulates a reactor loss-of-coolant accident with melted fuel in the center area and with heated, but unmelted, fuel toward the outer portions of the reactor core. Thus, in one fission-product-release experiment, the important effects of size and physical arrangement are simulated, permitting, to a degree, extrapolation to large-scale release.

Three of the seven-pin cluster experiments have been performed. Only the center pin was irradiated to serve as a fission-product source and to permit direct observation of the high-temperature plateau of fission products on the surrounding fuel. Purified helium flowed through the furnace (Fig. 3.7) and sampling system, as shown in the flow diagram, Fig. 3.9. The duration of heating was different for the experiments, resulting in 28% melting of the center pin in one experiment, 90% melting in

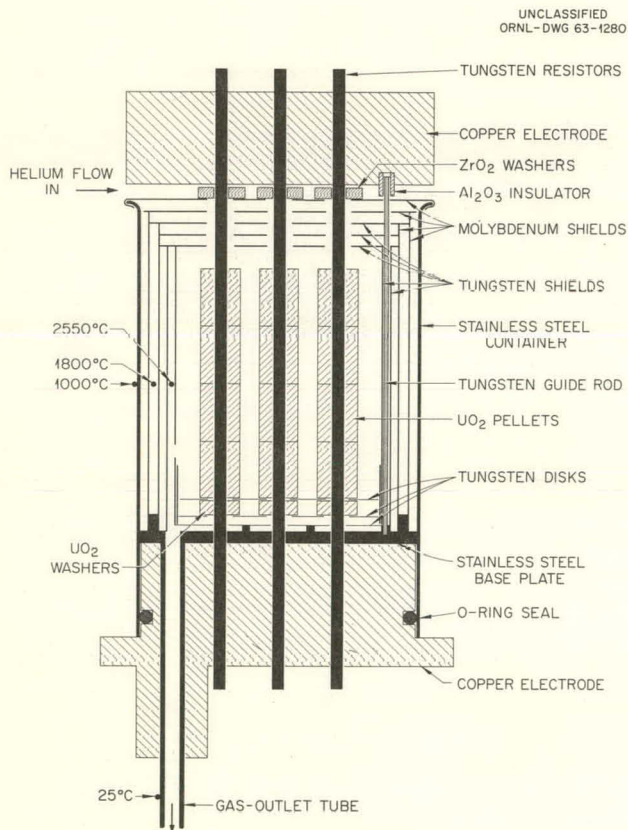


Fig. 3.7. Tungsten Resistor Furnace with Clustered Elements.

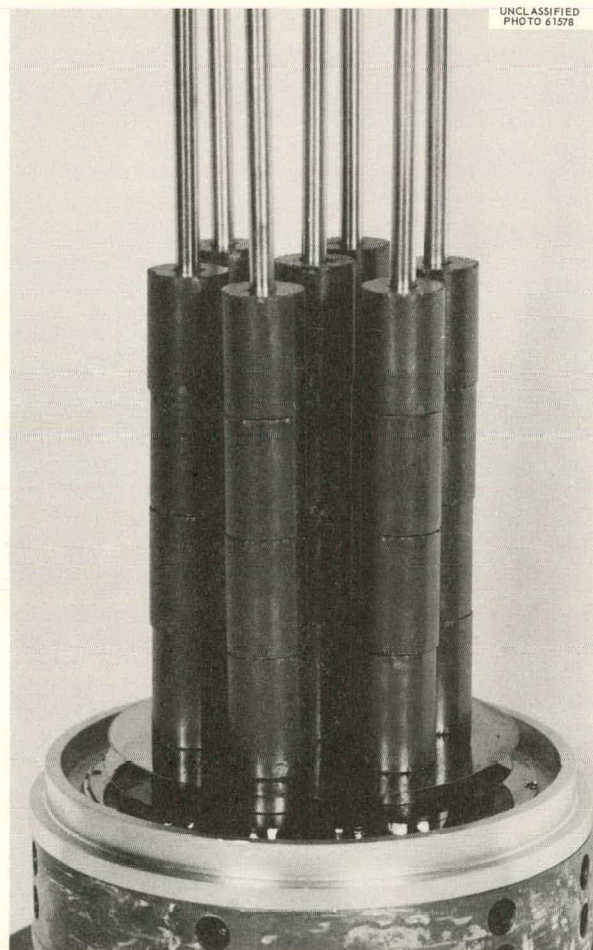


Fig. 3.8. Seven-Pin UO_2 Fuel Cluster Before Melting.

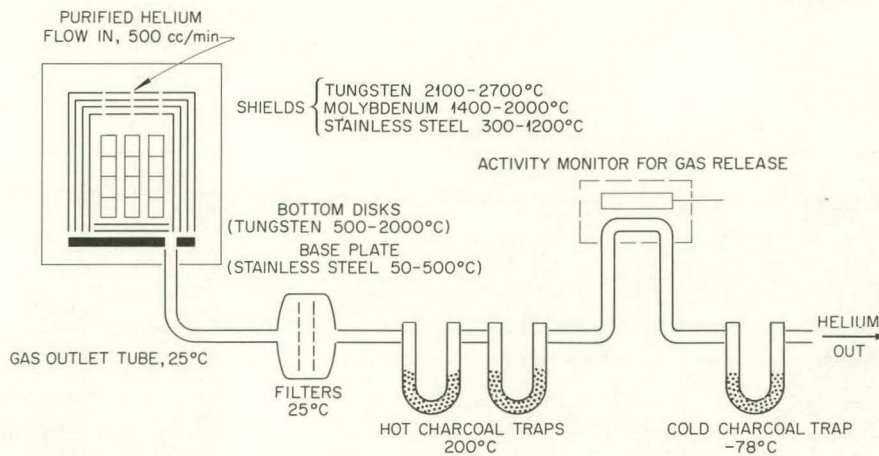


Fig. 3.9. Flow System for Fuel Pin Melting.

the second, and 60% melting in the third test, based on the fraction of xenon released from the tracer-level-irradiated center pins.

The extent of melting within the pins and the amount of molten UO_2 which flowed from the pins are indicated in Fig. 3.10. The outer surface of "skin" of the pellets in the 28%-melted experiment remained very nearly intact, as shown in Figs. 3.11 and 3.12. A few pieces of UO_2 broke off the outer-circle pellets. Some extrusion of UO_2 occurred through cracks in the side of center pellets, and molten UO_2 pushed aside a cracked section of the bottom pellet and poured out.

The experiment in which 90% of the center pin melted is shown in Fig. 3.13 after the shields were removed, and in Fig. 3.14 after the shell-like pieces of outer-circle pellets were removed. No loose pieces of the irradiated center pin were found.

Fission-product-release data obtained in the first two cluster experiments are shown graphically in Fig. 3.15. Total release values and the amounts found at each location are plotted against percent melting on a log-log scale. Total release values represent the sum of materials in all locations, except that plated out on unirradiated fuel. In general, fission-product release from these two experiments was directly proportional to the fraction of the fuel melting. Iodine, tellurium, and cesium were found in similar quantities in each location. Their concentration on the high-temperature shields was low, but they were the only isotopes found in significant quantity beyond the shields.

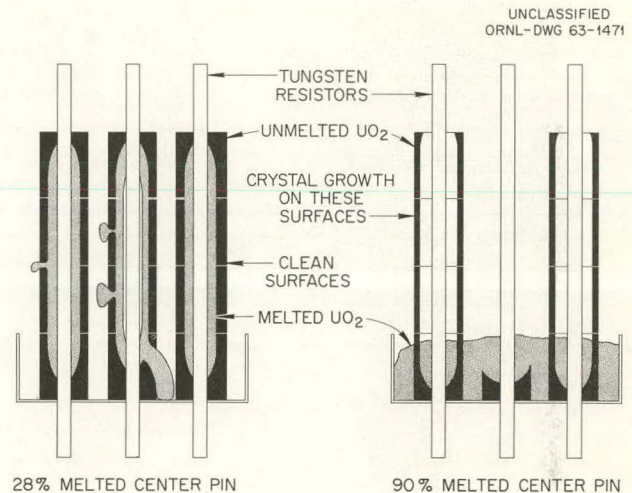


Fig. 3.10. Melting Pattern of Seven-Pin Fuel Cluster.

Comparison with Previous Single-Pin Experiments

Data in Table 3.6 permit a comparison of fission-product release from the seven-pin cluster experiments with release from single-pin experiments previously performed. The release numbers were corrected to 100% melting by direct proportion for each experiment, and the corrected values from comparable experiments were averaged. Release from the single-pin experiments was generally greater than that from the cluster experiments, with strontium and barium showing the greatest difference, although transport of these isotopes

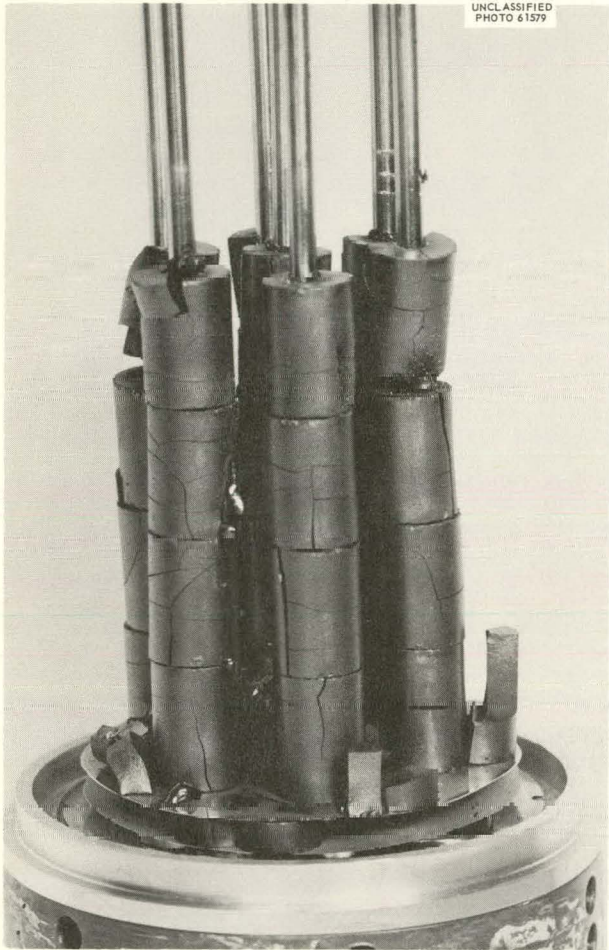


Fig. 3.11. Cluster After Melting Center Pin, 28% Melted.

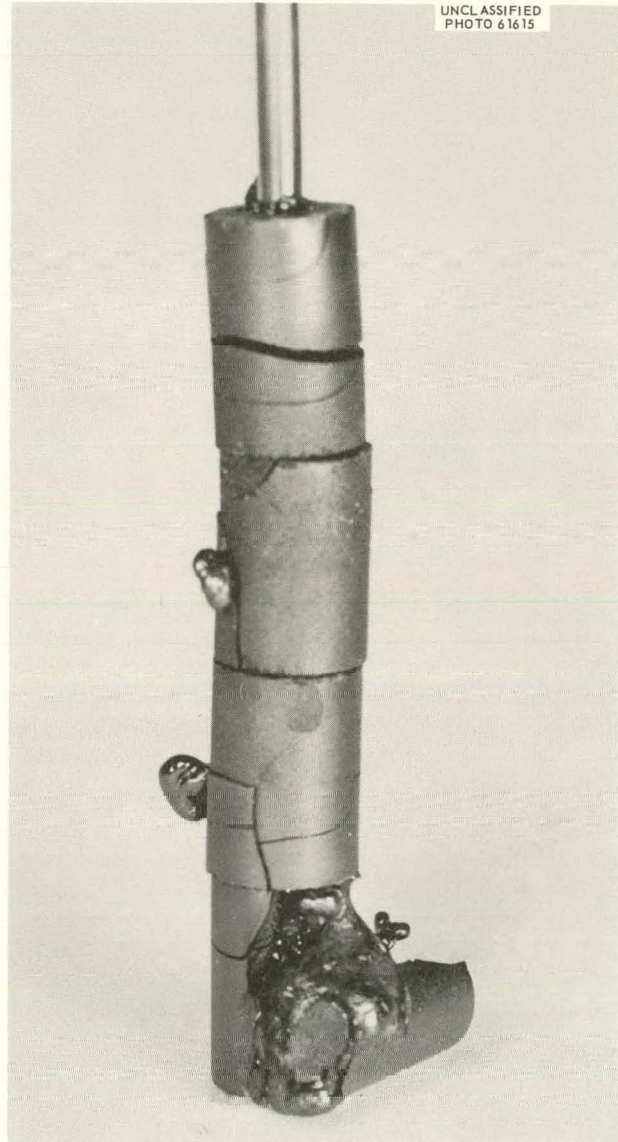


Fig. 3.12. Center Pin 28% Melted.

outside the furnace was very small. The higher release of ruthenium in the seven-pin cluster experiments was probably caused by some oxidizing agent present in the cluster arrangement.

The fraction of the I, Te, and Cs transported by the gas stream to the exit gas tube, filters, and charcoal trap from the seven-pin cluster was half that from the single-pin experiments, and the difference was even greater for UO_2 . The higher gas velocity through the smaller single-pin furnace may have caused the difference in transported release.

Plateout on Surrounding Fuel. — The amount of fission products plated out on the circle of unirradiated fuel was determined after melting by selecting samples of outer fuel pins with representative

gross-gamma activity, analyzing for fission products, and extrapolating from the sample surface area to that of the original full circle of six pins. The resulting amount of fuel plateout does not account completely for the difference in total release from the seven-pin and the single-pin experiments shown in Table 3.6. Since the single-pin and cluster experiments were conducted in different size furnaces, the effect of the outer fuel pins cannot be directly determined from the above comparison. Also, the poor precision obtained makes additional experiments necessary.

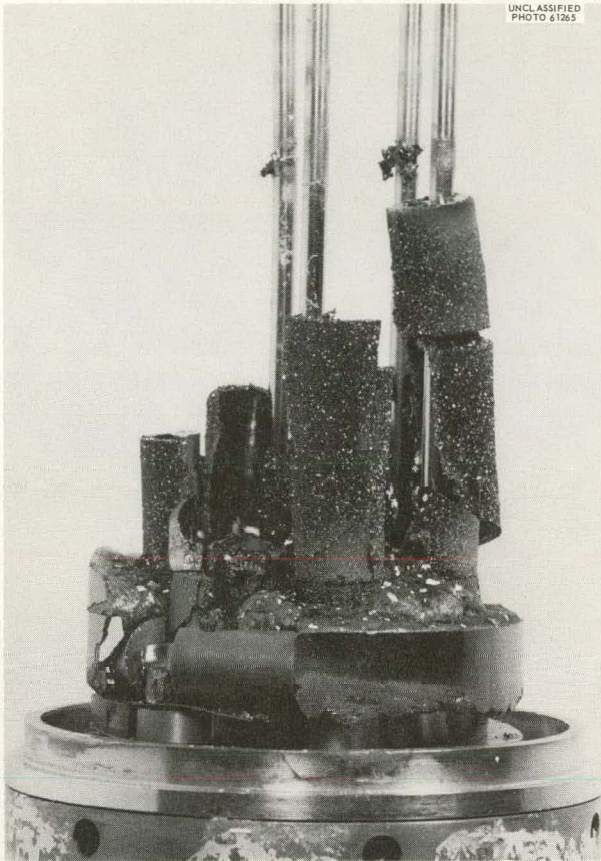


Fig. 3.13. Cluster After Melting, Center Pin 90% Melted.

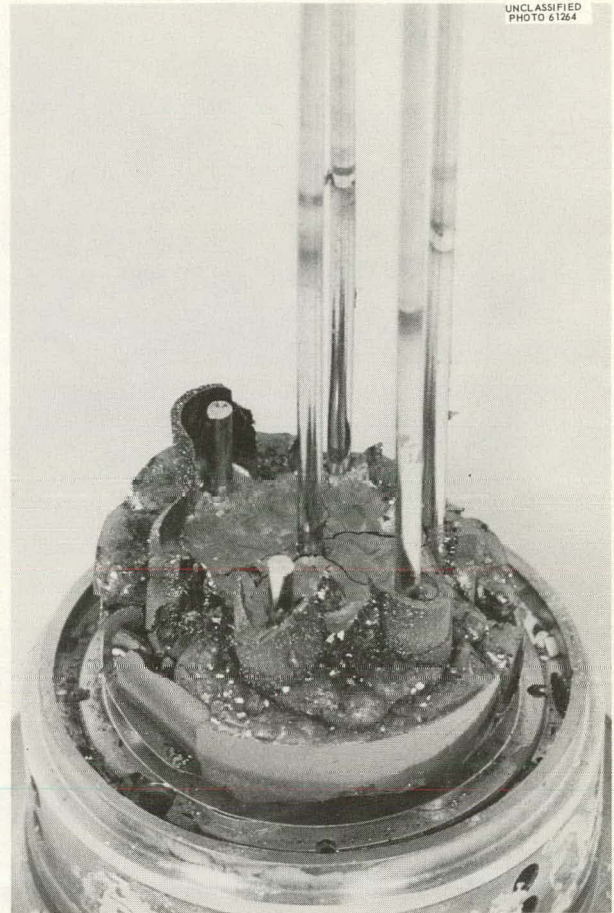


Fig. 3.14. UO₂ Puddle, Center Pin 90% Melted.

Material Transported Through a Restricted Gas Channel. — In the third experiment, performed with a seven-pin cluster arrangement, only three pellets per pin were used in a slightly different furnace from that employed in the previous experiments, as shown in Fig. 3.16. The bottom tungsten disk rested on the base plate and fitted snugly against the second cylindrical tungsten heat shield. This disk did not have the opening above the gas outlet tube which was present in all other single-pin and seven-pin furnaces. Thus, the helium leaving the furnace was forced to travel through the gaps around the bottom disk or outside the second

shield. The same helium flow rate was used as with the other seven-pin experiments.

The heating melted 60% of the center pin. Total fission-product release was approximately the same as from the other seven-pin experiments when corrected for the fraction melted. However, the amount of material transported to the gas tube, filter, and charcoal was considerably less, as shown by UO₂, I, Te, and Cs release data in Table 3.6. The large reduction in material transported out of the furnace is undoubtedly due to the longer and more intimate contact of exiting gas with shields, bottom disk, and base plate.

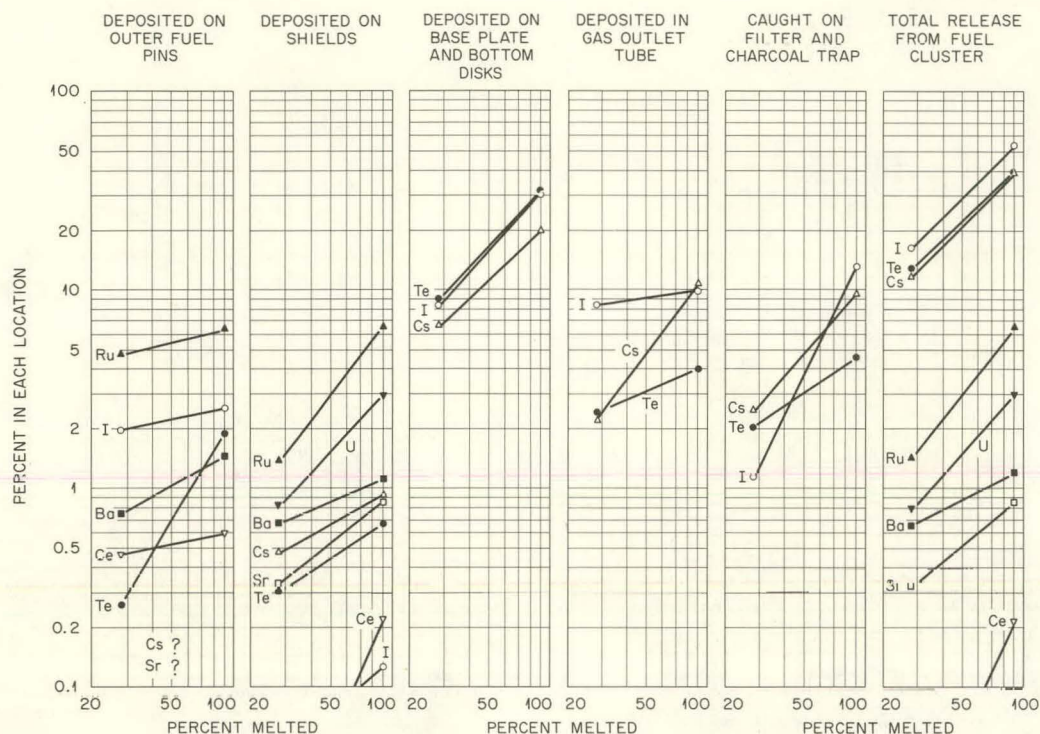


Fig. 3.15. Distribution of Fission Products Released from Partial Melting of the Center UO_2 Fuel Pin of a Seven-Pin Fuel Cluster in a Helium Atmosphere.

Table 3.6. Release from Bare UO_2 Fuel Pins Corrected to 100% Melting

Release Zone	Experiment Type	Release (%)							
		UO_2^a	I	Te	Cs	Ru	Sr	Ba	Ce
Total release from fuel cluster	1-pin ^b	0.81	70	90	82 ^c	1.1	2.5	9.0	<1.0
	7-pin ^d	3.1	60	45	37	6.3	1.1	1.9	0.12
Deposit on outer fuel pins	7-pin ^d		4.6	1.5		12		2.1	0.7
Transported release (gas tube, filter, charcoal)	1-pin ^c	0.003	52	40	31	<0.05	0.01	0.03	<0.1
	7-pin ^d	0.0002	28	13	19	<0.07	<0.0014	<0.0033	<0.002
	7-pin, restricted gas channel ^e	0.0003	1.5	0.67	<3.1				
Transported release to filter and charcoal	1-pin ^b	0.0008	25	17	21	<0.05	<0.003	<0.01	<0.004
	7-pin ^d	0.0001	9.4	6.3	9.8	<0.03	0.0007	<0.0003	<0.001
	7-pin, restricted gas channel ^e	0.00003	0.53	<0.21	<1.2				

^aBased on UO_2 in center irradiated pin; all irradiations were trace level (10^{14} fissions per gram of UO_2).

^bAverage of three experiments, three-pellet pin (39 g of UO_2), 360 cc(STP)/min purified helium, 0.67 fpm (2.9 volume changes per min) in furnace at 25°C.

^cData from one experiment in note (b).

^dAverage of two experiments, four-pellet pins (52 g of UO_2), 450 cc(STP)/min purified helium, 0.43 fpm (1.5 volume changes per min) in furnace at 25°C.

^eOne experiment, three-pellet pins (39 g of UO_2), 450 cc(STP)/min purified helium, 0.43 fpm (1.9 volume changes per min) in furnace at 25°C.

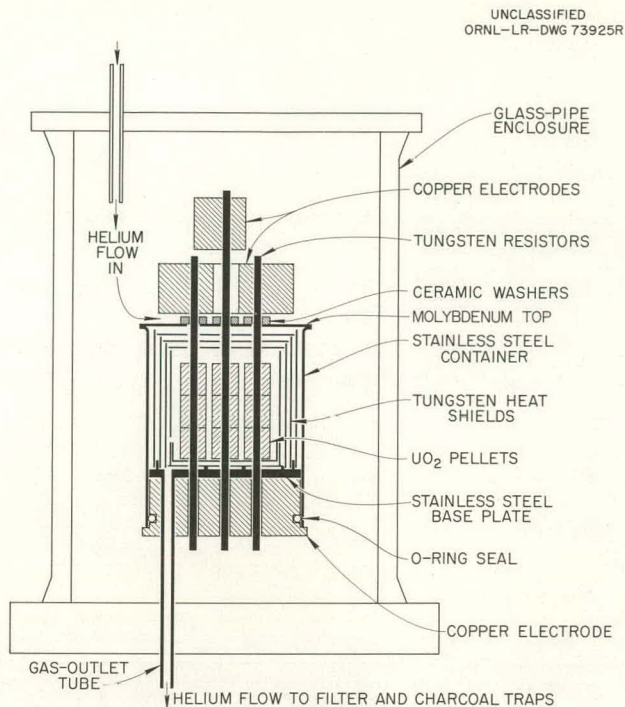


Fig. 3.16. Tungsten Resistor Furnace with Clustered Elements; Test No. 2-6-63.

EFFECT OF TIME AND GAS VELOCITY ON DISTRIBUTION OF FISSION PRODUCTS FROM UO₂ MELTED IN A TUNGSTEN CRUCIBLE IN HELIUM

Data on the effect of varying the time that tracer-level-irradiated UO₂ was held in the molten state in a helium atmosphere on the release of fission products were reported previously.² The time range for the period in the molten state (ref 2, Table 3.1) was too narrow to permit more than tentative conclusions concerning the effect of this variable. Consequently, additional experiments

²G. W. Parker et al., *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, p 9.

were performed with the same apparatus described in the previous report and with other conditions the same, except for time and gas velocity. The data obtained are shown in Table 3.7. Considering first the data obtained in experiments in which the gas velocity was constant and the time molten varied, it appears that the release of iodine and tellurium was virtually quantitative after 5 min in a low stream of helium, while cesium and barium release increased over the time range covered in these experiments. The fractions of the released iodine and tellurium found in the various parts of the apparatus showed no well-defined trends with varying time in the molten state, but the fraction of the cesium and barium in the furnace tube tended to increase with increasing time molten. Comparison of release results for experiments in which the time molten was virtually constant (4 to 5 min) shows that increasing gas velocity did not consistently increase the fraction of the released fission products transported outside the furnace tube, except for cesium and tellurium. In all experiments, a comparatively small fraction of the iodine released reached the charcoal trap, indicating that it may have been released in a low-volatility form or that it was adsorbed on large particles. Previously reported data² on the size of particles vaporized from molten UO₂ (0.03 to 1.0 μ) seem to indicate that the released iodine was not attached to particles of this type in these experiments. The upper end of the quartz furnace tube was relatively cool because of the heating method employed in these experiments and provided a convenient iodine deposition point, but transport of iodine to the furnace tube wall would not be expected to be independent of gas velocity as it appeared to be in these experiments. The amount of fission products remaining in the thorium oxide heat shield was determined in only a few experiments, but in those experiments in which separate analyses of the quartz furnace tube and the heat-shield leach solutions were performed, the iodine found was only a small part of the total amount in the furnace tube. Further experimentation is needed to clarify the apparently anomalous behavior of iodine in these experiments and to determine the chemical form of the fission products released in helium. It should be kept in mind that the UO₂ material employed in these studies was irradiated at tracer level, and previous experience indicates that caution must be employed in extrapolating tracer-level data to high-burnup conditions.

Table 3.7. Effect of Time in the Molten State and Gas Velocity on the Distribution of Fission Products Released from UO_2 in a Helium Atmosphere

Time Molten (min)	Helium Velocity ^a (fpm)	Fission-Product Location	Percentage of Fission-Product Inventory				Time Molten (min)	Helium Velocity ^a (fpm)	Fission-Product Location	Percentage of Fission-Product Inventory			
			I	Te	Cs	Ba				I	Te	Cs	Ba
1.5	2.4	Furnace	78.2	84.9	62.7	2.3	0.4	24	Furnace	22.1	57.2	29.8	0.7
		Chamber	7.7	2.0	10.0	0.1			Chamber	5.1	3.3	4.3	0.1
		Filters	1.3	4.9	7.5	0.2			Filters	15.0	22.8	17.0	0.7
		Charcoal	1.2						Charcoal	13.4			
		Total	88.4	91.8	80.2	2.6			Total	42.1	83.3	51.1	1.5
2.5	2.4	Furnace	69.6	85.4	73.2	2.9	1	12	Furnace	57.1	56.6	44.0	2.1
		Chamber	14.0	2.0	5.3	0.3			Chamber	4.0	0.9	0.3	
		Filters	8.2	8.8	10.9	0.4			Filters	21.9	26.9	19.4	0.4
		Charcoal	1.1						Charcoal	1.1			
		Total	92.9	96.2	89.4	3.6			Total	84.1	84.4	63.7	2.5
5.0	2.4	Furnace	40.3	78.6	83.1	3.3	5	12	Furnace	52.1	76.4	52.8	3.0
		Chamber	6.6	4.8	3.0	0.1			Chamber	6.5	1.0	1.0	0.2
		Filters	35.5	15.8	5.4	0.1			Filters	32.3	21.5	39.2	1.5
		Charcoal	17.2						Charcoal	5.9			
		Total	99.5	99.2	91.5	3.5			Total	96.8	98.9	93.8	4.7
10.0	2.4	Furnace	79.3	88.9	84.3	6.4	4	24	Furnace	38.4	52.1	32.2	3.5
		Chamber	6.8	2.6	3.0	0.1			Chamber	3.3	1.2	0.7	0.1
		Filters	8.9	7.8	7.7	0.1			Filters	25.3	38.4	9.0	0.2
		Charcoal	3.9						Charcoal	13.2			
		Total	98.9	99.9	95.0	6.6			Total	80.2	91.7	41.9	3.8

^aHelium flow measured at room temperature, and velocity calculated for the full furnace tube internal diameter.

4. Release of Fission Products on In-Pile Melting of Reactor Fuels Under Transient Conditions

G. W. Parker

R. A. Lorenz

Approval was obtained for ORNL fission-product-release studies in the TREAT facility at the NRTS following a review of the hazards analysis by TREAT management. The release of fission

products during transient melting, in an argon atmosphere, of a 10%-enriched UO_2 compact clad with type 347 stainless steel will be investigated in experiments conducted in this reactor. The

samples will be preheated to temperatures in the 800 to 1200°C range, and the released aerosol will be sampled by admission to an evacuated autoclave through a diffusion tube and filter shown in the flow diagram, Fig. 4.1. A more complete description of the program was included in the previous progress report.¹

The first two experiments have been assembled, leak-tested, and filled with argon. One of the

experiments with graphite spacing plugs and a container are shown in Fig. 4.2. Melting of the first few of the series at tracer irradiation level is scheduled in TREAT for late June.

¹G. W. Parker and R. A. Lorenz, *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, p 35.

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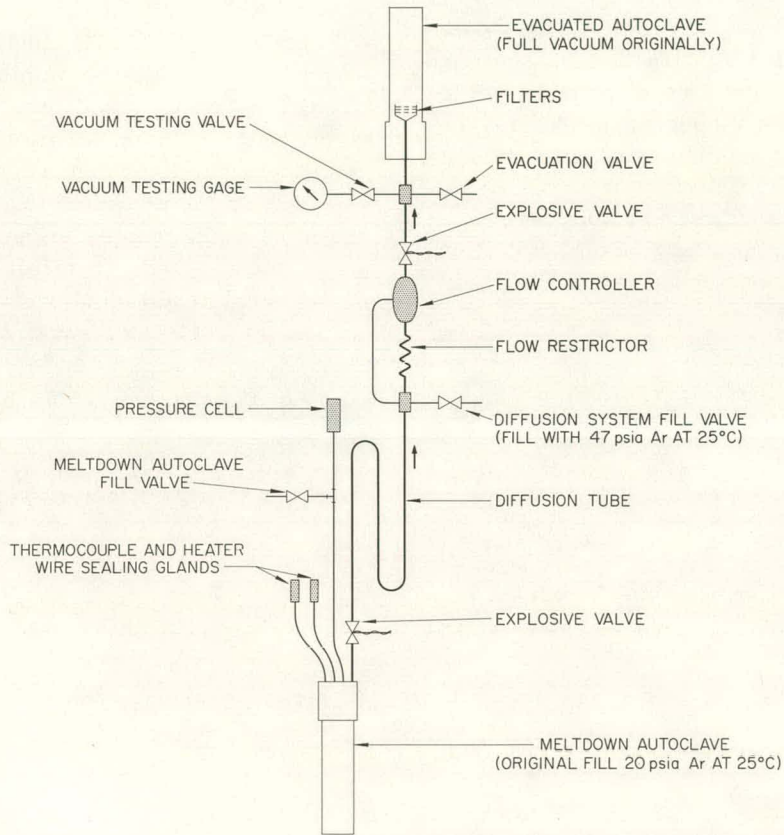


Fig. 4.1. Flow Diagram for Fission-Product Release Under Transient Reactor Conditions.

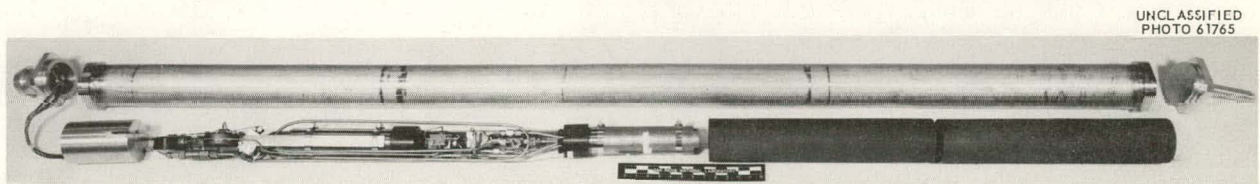


Fig. 4.2. Full-Scale TREAT Assembly and Graphite Spacing Plugs.

5. Release of Fission Products on In-Pile Melting or Burning of Reactor Fuels

W. E. Browning, Jr. R. P. Shields C. E. Miller, Jr. B. F. Roberts

In-pile experiments are being continued to study the release of fission products during simulated reactor accidents. Two types of experiments have been conducted in the ORR to simulate reactor accidents in which fuel elements were destroyed by melting or burning. One type consisted of melting or vaporizing a miniature stainless-steel-clad UO_2 fuel element in a helium atmosphere. In the other type, a miniature fuel element composed of spheroidal particles of uranium carbide, coated with pyrolytic carbon and embedded in a graphite matrix, was burned in air. In each case, fission and gamma heat raised the temperature of the fuel element high enough to cause destruction. The details of design and operation of these experiments have been described previously.^{1,2}

The analytical results of the first eight experiments have been discussed in detail,³ and the physical appearance of the UO_2 after the melting has been described.^{1,4}

UO_2 MELTING EXPERIMENTS

Experiments 9 and 10 continued the series in which UO_2 is melted in-pile under conditions simulating reactor accidents. The flow of helium sweep gas was increased from 400 to 800 cm^3/min for experiment 10; otherwise, the conditions were the same as the preceding UO_2 meltdowns. These experiments have been examined in the hot cell and analyzed radiochemically to determine the fission-product distribution. The results incorporated in Tables 5.1 and 5.2 indicate no marked

¹W. E. Browning, Jr., et al., *Nuclear Safety Program Semiann. Progr. Rept. June 30, 1962*, ORNL-3319, pp 29-38.

²W. E. Browning, Jr., et al., *Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1962*, ORNL-3262, pp 172-76.

³W. E. Browning, Jr., et al., *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, pp 27-35.

⁴W. E. Browning, Jr., et al., *Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1963*, ORNL-3417, pp 235-43.

Table 5.1. Material Released from the Fuel In UO_2 Melting Experiments^a

Experiment	Release (%)								
	Sr^{89}	Zr^{95}	Ru^{106}	I^{131}	Te^{132}	Cs^{137}	Ba^{140}	Ce^{144}	UO_2
2	99.8	87.3	88.0				99.7	93.0	50.0
3	93.0	86.9	51.2				96.1	85.2	42.7
4	76.6	55.5	82.8	99.6			94.1	58.9	44.5
5	57.3	42.9	90.8	99.4	98.4	97.2	55.4	44.6	34.5
6	54.6	60.1	59.8	99.0	91.4	98.6	51.7	48.7	46.8
7	5.21	0.908	10.9	96.0		96.5	14.0	2.50	0.710
8	43.7	58.1	57.4	98.7	97.6	98.0	70.0	43.4	36.0
9	71.3	71.5	93.1	99.7	99.1	98.8	71.2	70.3	70.5
10	68.8	57.7	95.3	79.3	95.9	94.9	72.5	59.3	55.8

^aSample length, 1 in.; diameter, 0.210 in.; sample molten 5 min except experiment 2, 10 min; helium flow, 400 cc/min.

Table 5.2. Material Released from High-Temperature Zone^a of the Furnace in UO₂ Melting Experiments

Experiment	Release (%)								
	Sr ⁸⁹	Zr ⁹⁵	Ru ¹⁰⁶	I ¹³¹	Te ¹³²	Cs ¹³⁷	Ba ¹⁴⁰	Ce ¹⁴⁴	UO ₂
2	20.1	0.115	2.84				14.5	0.508	4.08
3	1.24	0.110	0.614				0.732	1.78	0.0520
4	2.90	0.0465	6.60	94.5			0.941	0.651	0.773
5	1.44	0.0200	5.22	87.0	79.2	79.0	2.30	0.328	0.233
6	0.911	0.0896	1.32	94.3	48.8	81.9	0.547	0.0675	0.291
7	0.866	0.0370	2.15	49.7	11.5	74.1	0.576	0.322	0.00894
8	1.28	0.0508	1.73	95.4	81.4	87.5	0.824	0.244	0.0197
9	0.974	0.123	4.022	79.70	89.34	58.14	1.118	0.302	0.0128
10	2.119	2.819	0.122	89.46	71.45	79.73	2.837	1.003	0.289

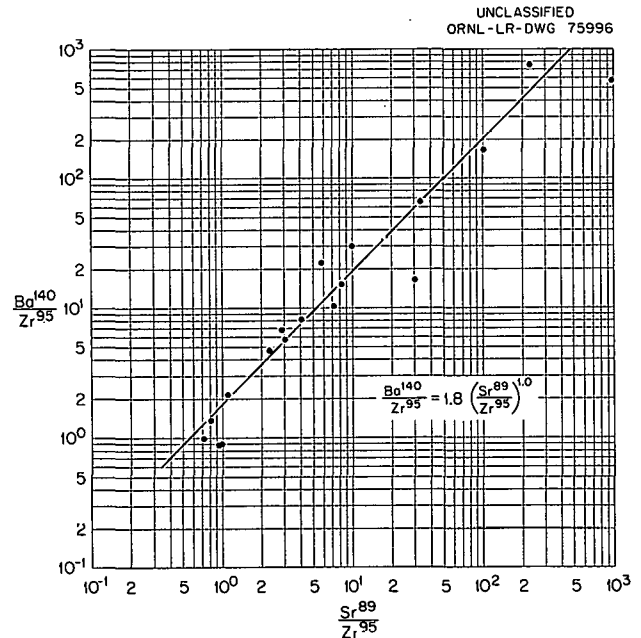
^aHigh-temperature zone includes fuel material and thermal insulation; minimum temperature, 1000°C.

change with gas flow rate, and, in general, previous conclusions regarding release have been substantiated. Data from earlier experiments are included in the tables for comparison.

CHARACTERIZATION STUDIES OF RELEASED FISSION PRODUCTS

The distribution of fission products and uranium among the various regions in each experiment is being analyzed and interpreted in terms of the fractionation processes which govern the behavior of these materials. Fractionation refers to any alteration of the radionuclide composition occurring during the experiment which results in a sample composition different from that predicted for normal fission yields. Similar fractionation studies have been reported by Freiling in a study of debris from nuclear detonations.⁵ Fractionation occurs because of differences in characteristics of the fission products that lead to differences in release or deposition mechanisms. A study of fractionation, then, should result in a better understanding of both the mechanisms and the characteristics. As an example of the type of data analysis which is being used in the fractionation studies, the ratio Ba¹⁴⁰/Zr⁹⁵ vs

Sr⁸⁹/Zr⁹⁵ is plotted in Fig. 5.1 for experiment 7. The equation of the line empirically determined is shown. In this plot the abscissa may be regarded as a measure of the intensity of the volatilization conditions to which each sample, represented by a point on the plot, has been

Fig. 5.1. Fractionation of Ba¹⁴⁰ vs Sr⁸⁹.

⁵E. C. Freiling, *Science* 3, 1991-98 (1961).

exposed. Since Sr^{89} is more volatile than Zr^{95} , points to the right represent samples with increasing amounts of volatilized Sr^{89} , for example, samples from cooler regions, while points to the left represent samples progressively depleted in Sr^{89} . The ordinate is a measure of the degree to which Ba^{140} volatilized for each volatilization condition on the abscissa. This plot indicates that Ba^{140} and Sr^{89} behave similarly and that both are more volatile than the reference Zr^{95} . In a similar plot for ruthenium from experiment 7, the slope is greater than unity, indicating that the ruthenium was more volatile than the strontium. In experiment 8 the slope for ruthenium was unity, indicating that its volatility was nearly the same as Ba^{140} . The difference in slope probably represents a difference in release mechanism, since the fuel in experiment 7 did not melt extensively while that in experiment 8 did. One result of the fractionation studies is the conclusion that in several of these experiments ruthenium follows the stainless steel cladding as the cladding melts into a puddle and subsequently vaporizes to other areas of the assembly. The mechanism for this is not yet known. However, it is anticipated that this work should lead to the formulation of models which explain the observed distributions and which may provide a basis for predicting the behavior, especially that in a controlled system, of accident-released fission products under various conditions. A knowledge of fractionation mechanisms will aid in the recognition of the relative importance of the various factors which define a reactor accident.

The characterization of released fission products by the diffusion tube method is being continued. Early results obtained by use of this method were reported previously.³

IN-PILE BURNING OF UC_2 -GRAPHITE FUEL

Three experiments have been conducted in the ORR at ORNL in which fuel specimens consisting of pyrolytic-carbon-coated uranium carbide particles embedded in a graphite matrix were burned at temperatures up to approximately 1400°C . In two of these experiments, fission and gamma heat produced a sufficiently high temperature for

a large portion of the fuel to burn during the 15-min period in which air was supplied as the sweep gas. The method of performing experiments and the results of the first experiment were described previously.³ The specifications for the specimen and pertinent experimental conditions are listed below:

Fuel specimen	
Particles	Uranium carbide spheres 175 to 205 μ diam, laminar pyrolytic-carbon coating 75 to 100 μ thick
Matrix	Graphite
Manufacturer	Minnesota Mining and Manufacturing Company
Dimensions	0.6-in. bushing, 0.5 in. long with 0.25-in. axial hole
Uranium content	8 to 10 wt % natural uranium
Burning conditions	
Initial temperature	890°C
Combustion temperature	Up to 1400°C
Air flow rate	400 cm^3/min
Burning duration	15 min

As the graphite matrix burned, some of the fuel particles fell away from the unburned fuel. Metallographic examination of these particles and those in the unburned fuel showed that the damage to particles ranged from none to complete destruction of the coating and oxidation of the uranium carbide; however, it was not possible to determine the percentage of particles which belong in these categories. Some of the particles which remained in the unburned fuel also had cracked coatings. In experiment 1, a count of the damaged and undamaged particles in the exposed surface of the fuel showed that about 45% of the observed particles had at least one crack in the coating. Experiment 2 was discarded without obtaining radiochemical analyses because the low uranium content of the specimen made a comparison with the other burning experiment impossible.

The results of experiment 3, for the most part, confirmed the conclusions from experiment 1. In the first experiment,^{3,6} 58% of the specimen burned as compared with 41% in the third. In the third experiment, most of the uranium carbide was in one side of the specimen, and this side burned very little; 93% of the uranium found by analysis was in the unburned fuel.

Fission-product-release data are shown in Table 5.3. The results of experiment 1, previously reported, are included for comparison. A large fraction of some of the fission products was probably retained in particles with undamaged pyrolytic-carbon coatings, especially those in the unburned fuel. The release of strontium, zirconium, barium, cerium, and uranium from the high-temperature zone of the furnace was very low, but large fractions of iodine, tellurium, and cesium were released. Forty percent of the ruthenium was released from the furnace in experiment 1 and 5.7% in experiment 3. The loss of ruthenium in the first experiment was probably due

⁶W. E. Browning, Jr., *et al.*, *Gas-Cooled Reactor Monthly Progr. Rept. September 1962*, ORNL-3372, pp 256-59.

to the formation of a volatile oxide. The determination of the release of krypton and xenon was omitted in order to avoid collecting oxygen in liquid-nitrogen-cooled charcoal adsorbers. The following conclusions, based on experiments 1 and 3, can be made:

1. Strontium, zirconium, barium, and cerium are retained where they are formed.
2. The unburned fuel retained 88 to 97% of the iodine associated with the uranium of the unburned fuel.
3. The following were released from the high-temperature zone of the furnace: 15 to 25% of the I, 15 to 25% of the Te, and 35 to 40% of the Cs.
4. The release of ruthenium varied between 5 and 40%.
5. An appreciable amount of Cs (10% of total), Ru (1%), and I (1.5%) penetrated the filter, but Te was retained by the filter.
6. The fragments which fell away from the specimen during burning contained the same percent of carbon as the unburned fuel.
7. The side of the specimen with the least amount of uranium carbide burns more rapidly than other portions.

Table 5.3. Distribution of Fission Products and Uranium in In-Pile Burning Experiments

	Material Found (Percentage of Total in Assembly)								
	Sr ⁸⁹	Zr ⁹⁵	Ru ¹⁰⁶	I ¹³¹	Te ¹³²	Cs ¹³⁷	Ba ¹⁴⁰	Ce ¹⁴⁴	U
Unburned fuel ^a									
Experiment 1	50.6	58.2	27.0	59.7	47.6	38.1	51.7	54.8	61.9
Experiment 3 ^b	75.4	83.9	87.0	82.4	76.1	39.1	84.9	85.3	93.9
Outside of fuel residue ^c									
Experiment 1	13.8	3.7	48.2	28.9	42.7	51.3	14.5	9.05	2.12
Experiment 3	13.1	5.04	8.81	16.5	23.2	54.5	6.96	6.98	0.877
Retained in furnace									
Experiment 1	98.8	99.5	59.3	73.2	64.9	63.0	99.5	93.8	99.6
Experiment 3	98.7	98.7	94.3	85.6	85.7	58.4	99.3	97.1	99.8

^a58.6% of the sample was burned in experiment 1 and 41% in experiment 3.

^bExperiment 2 was not analyzed because of the low uranium content of the specimen.

^cFuel materials on all exposed surfaces except the unburned portion of the specimen, fuel particles and powder, and the floor of the combustion furnace.

The results of melting and fuel-burning experiments included in this report confirm the following previously reported conclusions.³ Although large fractions of the fission products were released from the fuel, they were almost entirely retained inside the high-temperature zone of the furnace with the exception of iodine, cesium, and tellurium; when oxygen was present, ruthenium was also released. The noble gases appeared to be released completely. These results suggest that,

for the conditions tested, effective retention of many fission products may be expected by high-temperature surfaces near the hot zone in reactor accidents. Of the fission products released from the high-temperature zone, significant fractions were carried as particles as small as 20 to 30 Å in diameter. This work is being continued with emphasis being placed on the effects of the conditions of reactor accidents on the amounts and forms of released fission products.

6. Characterization and Control of Accident-Released Fission Products

W. E. Browning, Jr.

R. D. Ackley

CHARACTERIZATION OF RADIOACTIVE IODINE COMPOUNDS BY DIFFUSIONAL DEPOSITION

The development of a method of characterizing gas-borne radioactive vapors and particulate matter and its application in the study of the removal of such materials from gases was previously described.¹ In this method, diffusion coefficients, which are related to molecular properties or to particle size, are determined by measuring the distribution of radioactivity deposited on the walls of a channel as a result of exposure to gas which carries radioactive materials and which flows under laminar conditions. This technique was also utilized in an effort to identify compounds of iodine, whose presence became apparent in recent experiments.

In the earlier work, the diffusion-channel technique was used to investigate, on a laboratory scale, the behavior of millimicron-size particles carrying radioiodine in a variety of gas-cleaning systems. In later experiments, diffusion-tube deposition profiles were obtained which did not conform to expected behavior; for example, there

were pronounced maxima at a considerable distance from the tube entrance that required a complex mechanism involving iodine compounds to explain them. To eliminate the possibility that the spark gap used in generating particles was a factor in this effect, further experiments were performed in which deliberate particle generation was omitted and the source air was carefully filtered. Examination of the silver-plated copper diffusion tubes, associated tubing, and activated carbon traps indicated that two distinct species of iodine other than the elemental form were present. One species appeared to be quantitatively retained by the inner surface of rubber tubing and the other by activated carbon. Neither was quantitatively retained by silver metal. A somewhat similar experiment, in which particle generation was omitted, was performed in this laboratory about a year earlier; at that time no significant indication of these compounds was observed. British workers in this field have reported that their data indicated some formation of iodine compounds when elemental iodine is released into atmospheric air.² Their results, however, were obtained with considerably lower

¹W. E. Browning, Jr., and R. D. Ackley, *Nuclear Safety Program Semiann. Progr. Rept. June 30, 1962*, ORNL-3319, pp 44-50; *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, pp 44-51.

²A. C. Chamberlain *et al.*, *Discussions Faraday Soc.* 30, 162-69 (1960).

concentrations of iodine in air and so are probably not directly comparable with the results of the present experiments.

The presence of the two as yet unidentified iodine species referred to above may be a result of impurities in the elemental iodine source or in the air source, or a result of reaction with materials in the apparatus, and may or may not be of general concern. More explicitly, the two species may be singular to the present experiments or they may tend to occur also in other laboratory work; an additional and more serious possibility is that the compounds may be produced when iodine is released into some or all reactor environments. In any event, considering the important role of iodine in nuclear safety, elucidation of the origin and identity of each of the two species appears to be a highly desirable objective, since such information is needed to interpret laboratory work properly and may also be needed to permit the design of adequate gas-cleaning systems for nuclear reactors.

One approach employed was an attempt to determine accurately the diffusion coefficient of

the compound which readily reacted with a rubber surface. The experimental arrangement is shown in Fig. 6.1. The iodine was introduced into the main air stream flowing at a rate of 3 liters/min so as to give a concentration of about $10^3 \mu\text{g}/\text{m}^3$. The quartz tube, at 25°C , provided about 6 sec of interaction time for the iodine and air. Diffusion tubes DC1 and DC5 were included to permit comparison with previous experiments. The squalene-coated tube, DC2, was used to present a surface which might behave similarly to rubber, but it did not yield useful results. The tube with activated carbon, DC4, indicated essentially complete retention of the two compounds by about a 3-in. depth of activated carbon at room temperature with an air velocity of about 4 fpm. Results obtained with a composite diffusion tube, with first silver and then rubber inner surfaces, DC3, are shown in Fig. 6.2. An average of all the results obtained for deposition on rubber, including the $0.039 \text{ cm}^2/\text{sec}$ of Fig. 6.2, yields a diffusion coefficient for the compound in air of approximately $0.05 \text{ cm}^2/\text{sec}$.

To obtain an idea of what this diffusion coefficient of $0.05 \text{ cm}^2/\text{sec}$ implied, use was made of

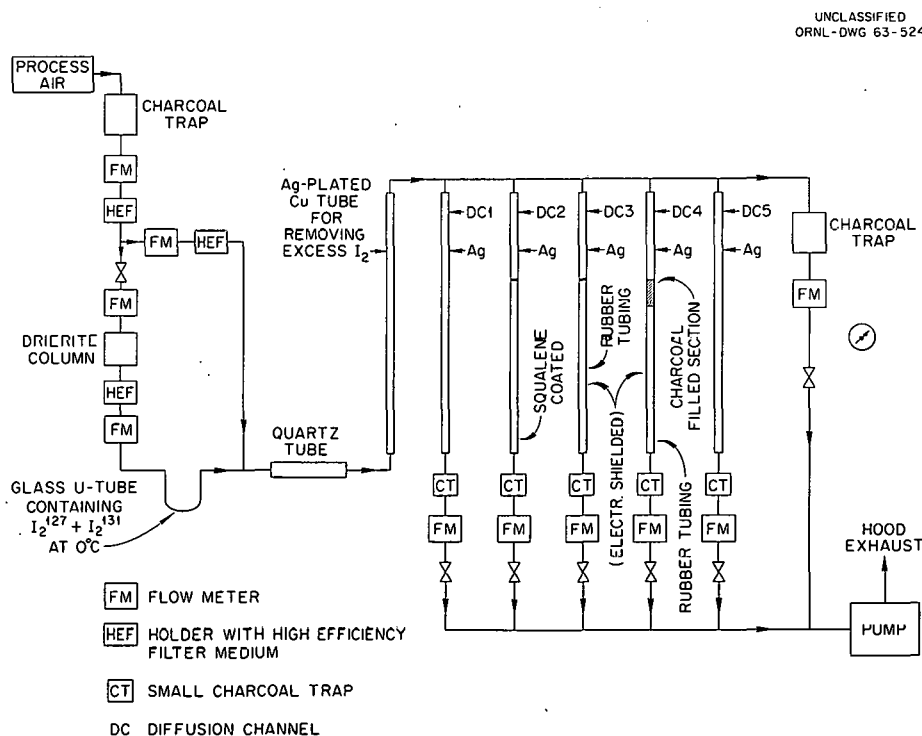


Fig. 6.1. Apparatus for Investigating Diffusional Behavior of Iodine Compounds.

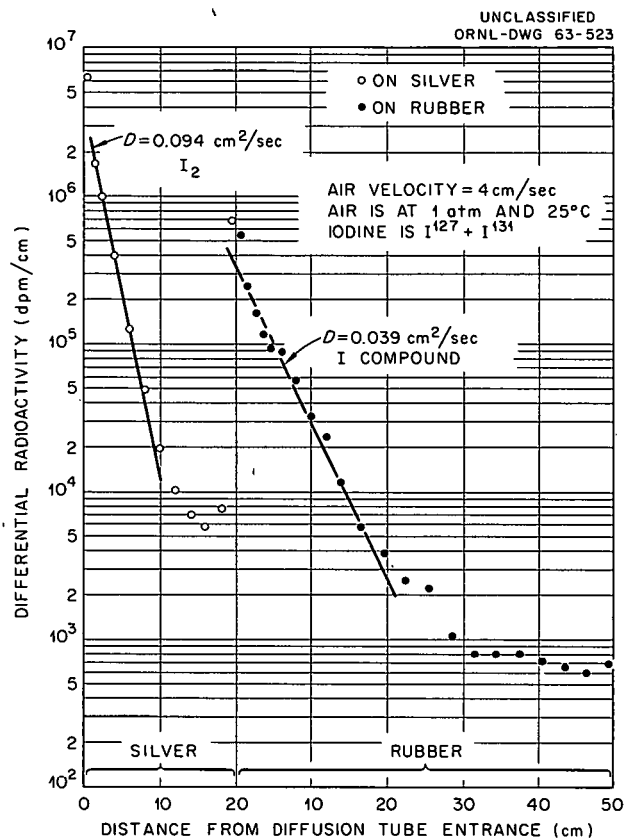


Fig. 6.2. Diffusional Deposition of Elemental Iodine and an Iodine Compound.

the Gilliland equation for estimating diffusion coefficients and also of Schroeder's method of estimating molal volumes at the boiling point because molal volumes are involved in Gilliland's equation along with molecular weights, pressure, and temperature.³ This procedure indicated that the observed diffusion coefficient could be explained by such compounds as dodecyl or tridecyl iodide, molecular weights equal to 296 and 310 respectively; however, this is not to suggest that the unknown compounds are either of these but rather to provide clues as to the molecular weight and size of the compound sought.

In another approach, the material remaining in the iodine U-tube at the conclusion of the experiment in which the data shown in Fig. 6.2 were

³R. C. Reid and T. K. Sherwood, *The Properties of Gas and Liquids*, pp 51, 52, and 268, McGraw-Hill, New York, 1958.

obtained was analyzed by mass spectrometry, and it was observed to contain two compounds with masses well in excess of that of elemental iodine, the masses being 284 and 338 (amu). Detailed analysis of the spectrum revealed that at least the compound of mass 338 contained an iodine atom, since mass values lesser by 127, the mass of iodine, were observed. Making the not unreasonable assumption that the molal volumes of the liquid at the boiling point of these compounds are equal numerically to their masses and using an assumed density of unity, application of the Gilliland equation yields 0.054 and 0.049 cm^2/sec respectively.

Thus, some progress has been made toward establishing the origin and identity of these compounds, and additional work with this objective is currently under way. Also, since there is some indication that the iodine source is responsible for the appearance of the compounds, a part of the effort is being directed toward alternate methods of producing elemental iodine for use in these studies.

MEASUREMENT OF CHARACTERISTICS OF RADIOACTIVE AEROSOLS USING FIBROUS FILTERS

The characterization of accident-released fission products, especially in the form of gas-borne particulates, is necessary so that effective methods may be developed for removing radioactivity from gas streams. The transport of an aerosol through an array of fibers brings into play the processes of inertial impaction, interception, and diffusion. Since all these processes have important effects on the behavior of radioactive materials in gases, it would be useful to be able to characterize radioactive aerosols by measuring their behavior in fibrous beds. A method has been developed for measuring aerosols in terms of their response to these processes by determining their distribution vs depth in fibrous filters under carefully controlled conditions.

The experimental techniques have been reported previously⁴ but will be reviewed briefly. A radioactive aerosol containing zinc particles ranging

⁴W. E. Browning, Jr., and M. D. Silverman, *Nuclear Safety Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, pp 50-55.

in diameter from 20 to 300 Å, tagged with Zn^{65} , is transported through a series of uniform Dacron filters. Radioassay is employed to estimate the particle distribution vs depth through the filter bed, which is composed of discrete layers of fibers.

A series of experiments has been conducted over a wide range of velocity (i.e., linear flow rates from 0.4 to 88 fpm). The relevant data obtained are shown in Fig. 6.3, where the fraction of activity collected is plotted against the filter depth, expressed as fiber length per unit of filter area. The slopes of these distribution curves are used to estimate how much of the filtration is accomplished by each of the major interaction mechanisms mentioned above.

The filtering action of a fiber mat results from the cumulative effect of particle removal by individual fibers. The collection efficiency of an individual fiber, η , is defined as the ratio of the cross-sectional area of the aerosol stream from which particles are removed to the projected area of the collector in the direction of flow. Fiber efficiencies have been calculated for the experiments conducted at linear velocities of 0.4 to 10 fpm (diffusion region) by use of the expression

developed by Chen:⁵

$$\eta_{\alpha} = -\ln \frac{N}{N_0} \frac{\pi \epsilon (d_f)_s^2}{4 \alpha L (d_f)_{av}}, \quad (1)$$

where

η_{α} = single-fiber efficiency in mat of fraction solids α ,

N_0 = upstream aerosol concentration,

N = downstream aerosol concentration,

ϵ = porosity (fraction voids),

$\alpha = 1 - \epsilon$ (fraction fibers),

L = thickness of mat,

$(d_f)_s$ = mean surface fiber diameter,

$(d_f)_{av}$ = average fiber diameter.

Because $(d_f)_s$ and $(d_f)_{av}$ are about equal for the fiber employed, the expression reduces to

$$\eta_{\alpha} = -\ln \frac{N}{N_0} \frac{\pi \epsilon (d_f)_{av}}{4 \alpha L}. \quad (2)$$

⁵C. Y. Chen, *Chem. Rev.* 55, 595 (1955).

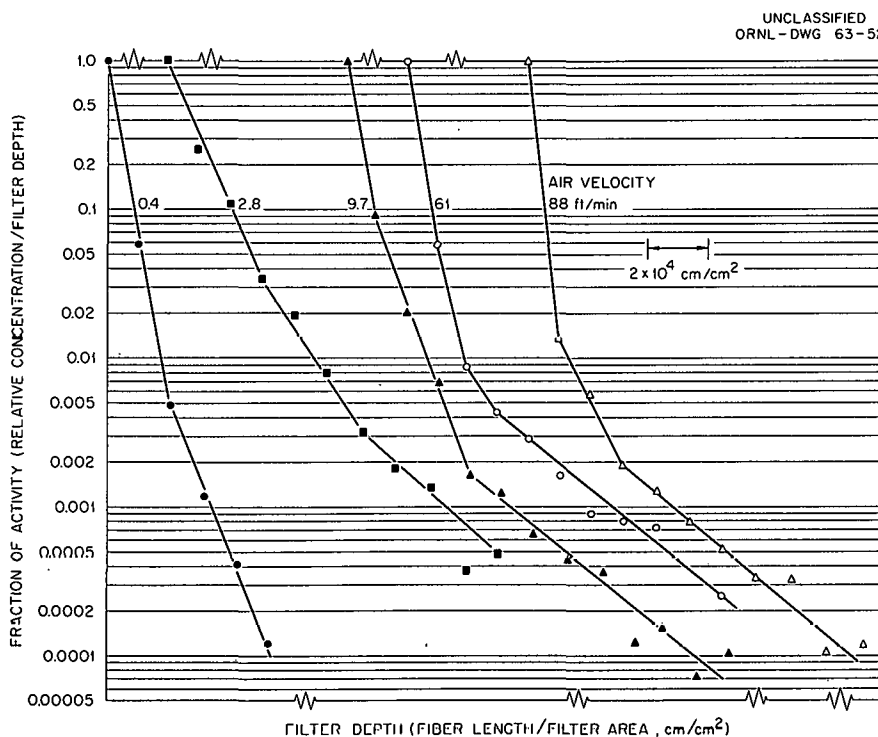


Fig. 6.3. Distribution of Zinc Activity vs Depth in Filter.

Figure 6.4 is a plot of the fiber efficiency vs the linear velocity; the inverse relation, $\eta_a \sim (1/\text{velocity})^x$, shows that diffusion is the primary mechanism in the filtration of the Zn⁶⁵ aerosol.

Stairmand,⁶ using the same approach as Langmuir,⁷ based on particles diffusing to the collector surface in a quiescent fluid within a time equal to $\pi d_f/2v$, derived the following expression for the diffusion collection efficiency,

$$\eta = \left(\frac{8D}{vd_f} \right)^{1/2}, \quad (3)$$

where v is the gas velocity, d_f and d_p are the diameters of the fiber and particle, respectively, and D is the diffusion coefficient calculated from the Einstein expression:⁸

$$D = \frac{CkT}{3\pi\mu d_p}. \quad (4)$$

The Stairmand relation, Eq. (3), has been used to calculate particle diameters (column 2) from the data obtained for the series of experiments shown in Table 6.1. The double sets of figures represent the two groups of particle sizes, as

⁶C. J. Stairmand, *Trans. Inst. Chem. Engrs. (London)* 28, 130 (1950).

⁷I. Langmuir, Office of Scientific Research and Development Report No. 865 (1942).

⁸A. Einstein, *Ann. Physik* 17, 549 (1905); 19, 371 (1906).

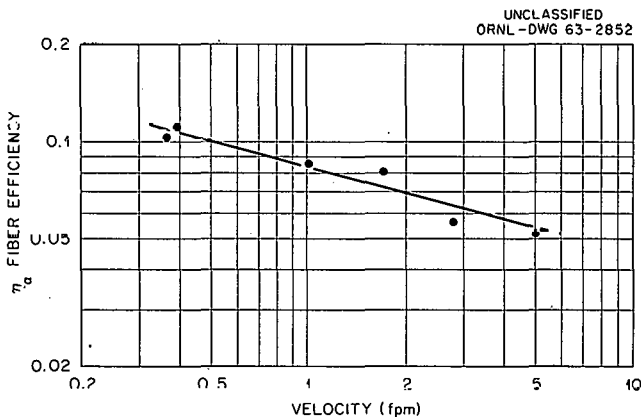


Fig. 6.4. Effect of Velocity on Individual Fiber Efficiency.

exemplified by the separate portions of the curves shown in Fig. 6.3. In the last column of the table under the heading "radiochemical data" are listed another set of particle diameters, estimated from the experimental data in the following manner. Radioassay results from the filter mats, along with specific activity data from the irradiated zinc foil electrodes, were combined to give the total weight of zinc removed by the filters in each experiment. The number of particles produced was calculated from the volume of air transported and a particle concentration for the aerosol (read from a condensation-nuclei counter, Gardner Associates). The concentrations observed are consistent with those which would be expected from the geometry of the experiment and coagulation theory expounded by Whytlaw-Gray.⁹ Knowing the weight of aerosol filtered and the number of particles per experiment for the particles in each experiment. It should be recognized that the latter diameters are weighted toward the larger particle sizes, since more

⁹R. Whytlaw-Gray, *J. Chem. Soc.* 1935, 273 (1935).

Table 6.1. Particle Sizes Determined from Filtration Experiments

Linear Velocity (fpm)	Particle Diameter (A)	
	Calculated by Use of Eq. (3)	Radiochemical Data
0.4	105	120
	196	
0.4	103	150
	140	
0.7	100	135
	180	
1.0	52	135
	98	
1.7	36	120
	70	
2.8	35	90
	105	
5.0	13	90
	28	

radioactivity is associated with them. It should be noted also that at the highest flow rate shown (5 fpm) the particle diameters calculated theoretically are low compared with the radiochemical values, because the interceptional process is contributing to the filtration. The diameters listed in the table may be compared with the figures given in the particle distribution plot (Fig. 6.5) obtained from electron microscopy photomicrographs.

The agreement among the results obtained by use of three different methods of estimating particle size is quite good, indicating that these filters can be satisfactorily employed as a particle analyzer in the diffusion region. The data in the interceptional and inertial regions are now being studied to determine whether this technique will find similar application.

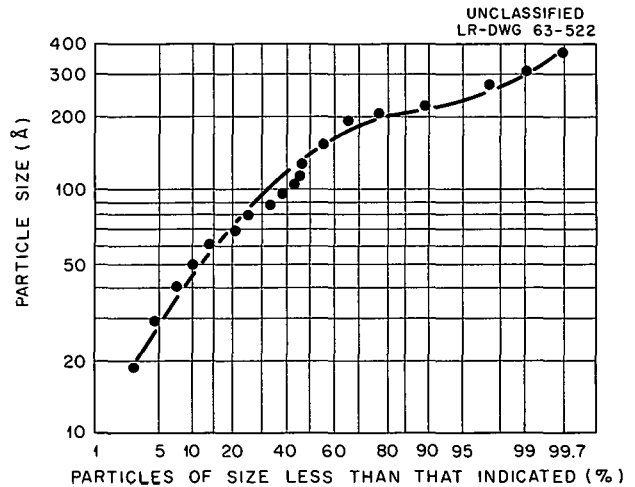


Fig. 6.5. Particle-Size Distribution of Zinc Oxide Particles.

7. Fission-Product Transport Evaluation

C. J. Barton

G. W. Parker

G. E. Creek

W. J. Martin

DISTRIBUTION OF FISSION PRODUCTS RELEASED FROM MOLTEN ALUMINUM-URANIUM ALLOY

A systematic analysis of the location of the individual fission products released in three atmospheric environments, air, helium, and an 80% steam-20% air mixture, is shown in Table 7.1. This type of study is of value as a measure of the transport efficiency and, in the case of the steam atmosphere, of the washout effect resulting from condensation of steam. A comparison of the apparent chemical effect of each atmosphere is also of interest, particularly as it affects iodine and tellurium. The data show that up to 88% of the iodine released in air was apparently in the molecular form and it was carried through the filters to charcoal absorbers. In helium, on the other hand, the percentage of iodine reaching the charcoal is 0.1% or less with the bulk having deposited in the furnace tube and on the absolute

filters, indicating that almost all the iodine was in the reduced form. Tellurium also showed a different behavior in the helium atmosphere. Since very little release occurred from the residual metallic fuel, oxidation evidently has a significant effect on the volatility of tellurium from the alloy; however, the fact that little or no ruthenium was released even in air or steam atmospheres suggests that tellurium is much more easily oxidized under the conditions prevailing in these experiments.

Steam washout accounted for up to 75% of the iodine; even with 20% by volume of air present in the saturated steam, only 10% of the iodine remained volatile enough to reach the charcoal. Cesium and tellurium were less effectively removed by steam condensation than iodine. In general, the fraction of each nuclide transported as far as the filters was below 50% and often as low as a few percent, except for iodine in oxidizing atmospheres.

Table 7.1. Transport of Fission Products Released from Uranium-Aluminum Alloy

Transport Fraction	Temp	Iodine			Tellurium			Cesium			Ruthenium		
		Air	Helium	Steam Air	Air	Helium	Steam Air	Air	Helium	Steam Air	Air	Helium	Steam Air
Total release	700	37.8		27.0	0.3		0.03	3.1		0.62	0.02		0.02
(%) at total inventory	800	78.6	29.8	76.8	0.2	5.3	0.26	3.8	13	1.1	0.1	0.18	0.1
	900	91.9	52.8	90.6	2.1	4.3	5.65	6.2	20.8	6.5	0.1	0.08	0.45
	1000	97.3	82.1	95.6	9.7	2.8	22.6	8.8	47.7	11	0.23	0.19	0.5
	1100	98.4	82.4	96.8	44.8	2.2	67.9	12.4	69.5	30.5	0.6	0.025	0.8
	1150	94.2			62.0			18.6			0.36		
Percent of total inventory plated on hot zone	700	19.9		17.0	0.3		0.03	3.05		0.5	0.02		0.02
	800	30.6	28.4	9.3	0.2	5.3	0.25	3.75	12.9	0.74	0.1	0.18	0.1
	900	11.3	51.3	12.9	1.9	4.3	3.68	5.94	20.6	4.3	0.1	0.08	0.4
	1000	38.2	69.6	13.0	9.6	2.8	12.6	8.06	45.8	6.86	0.2	0.18	0.5
	1100	6.3	65.8	10.3	37.9	2.2	22.1	11.5	63.1	14.9	0.5	0.02	0.3
	1150	17.4			40.4			17.0			0.35		
Percent of total inventory transported to filters by particulates	700	2.83		0.32	0.02		0.0002	0.05		0.02			0.0002
	800	4.5	1.4	0.14	0.04	0.002	0.006	0.05	0.07	0.048	0.0001	0.004	0.001
	900	4.2	1.5	0.7	0.2	0.004	0.2	0.26	0.16	0.25	0.006	0.001	0.006
	1000	0.9	13.3	4.1	0.04	0.017	1.17	0.73	1.9	0.43	<0.05	0.009	0.0045
	1100	3.9	17.4	3.9	6.9		5.7	0.89	6.3	1.65	0.05	0.003	0.42
	1150	9.2			21.6			1.6			0.05		
Percent of total inventory absorbed in charcoal traps (molecular inventory)	700	15.0		0.002									
	800	44.2	0.08	0.05									
	900	76.4	0.03	0.7									
	1000	59.0	0.11	8.5									
	1100	88.0	0.06	9.6									
Percent of total inventory collected in the steam condensate	700			9.8			0.0001			0.9			0.0001
	800			66.9			0.03			0.31			0.02
	900			77.0			1.77			1.95			0.02
	1000			69.8			8.7			3.71			0.01
	1100			73.0			40.0			13.96			0.08

HOT-CELL CONTAINMENT MOCKUP FACILITY FOR TRANSPORT EVALUATIONS

A major uncertainty in the application of the results of small-scale release experiments in reactor hazards analyses is the efficiency of transport from the melting site (the reactor core) to the containment system, usually a large pressure shell around the reactor. In the absence of reliable data on the magnitude of efficiency factors, the designer must assume 100% efficiency and thereby accepts a heavy penalty in his hazards analysis. In order to provide some knowledge of transport coefficients, a definitive experiment must simulate both the reactor failure configuration and also the factors which affect assisted or unassisted plateout of fission products which are inherent or normal to the reactor. The important parameters of burnup, atmosphere, temperature, etc. must also be included in the simulation, since active surface areas are likely to become saturated in proportion to the concentration of fission products. The eventual effect of larger concentrations of released fission products and other volatile components of the fuel material and its immediate environment on the properties of the released aerosol can only be speculated upon at this time, since no realistic experimental data are available.

A scaled-up hot-cell facility has been under design since the early days of the out-of-pile release program. Construction of this facility was recently undertaken to provide for the handling of multicurie fuel capsules which represent an increase in sample size by at least two to three orders of magnitude over that presently handled in the laboratory hood facility.

The design of the facility has been changed from that originally conceived in order to permit study of transport phenomena and to provide for more precise particle-size fractionation of the smoke and fumes. Shown in Fig. 7.1 is the current model of the Containment Mockup Facility. The apparatus is designed to permit the fission-product cloud to be transferred as quickly and directly as possible into the containment chamber. Here it may be delayed for a definite period of time for the evaluation of the agglomeration, plateout, and washout effects before the residual air-borne material is displaced through filters and iodine-absorption beds.

The Hot-Cell Containment Mockup Facility has an added feature, a mockup test filter holder which has diffusion tubes connected to it, as shown by Fig. 7.1. This arrangement will permit the measurement of the filterable air-borne solids, as well as the less than $0.01\text{-}\mu$ size particles which do not collect efficiently on the absolute filters. The components of the mockup filter will be duplicates of the commercial products proposed for large reactors.

The facility will be used first in the appraisal of the plateout and steam washout effect on highly irradiated aluminum alloy. A short section of $\frac{1}{16}$ -in.-ID Transite air duct in the off-gas line to the filters has been included since it was thought that Transite might enhance deposition or absorption on the side walls. The small duct diameter was chosen to provide Reynolds numbers of the same magnitude as those established in reactor design criteria. To continue the high degree of simulation, the exact gas velocities proposed for the ORR containment filter system are also being included as experimental parameters.

In operation, a long sample of fuel metal plate will be suspended in the resistance-heated furnace tube shown below the mockup containment chamber. With the chamber at partially reduced pressure, the sample will be heated to melting, beginning at the bottom. The flowing metal will drip from this end, and the melt will collect in a crucible provided for that purpose.

An atmosphere of steam with about 10% air will pass over the specimen carrying the cloud into the chamber. After a given interval (10 min to 1 hr), the chamber will be filled to atmospheric pressure with the steam-air mixture. At this point, the contents of the chamber will be drawn through the filter to determine the residual amount of air-borne material. Finally, the furnace tube will be disconnected and subjected to chemical leaching, the metal residue will be dissolved, and the containment chamber will be leached to determine the fraction of the fission-product inventory that reached the containment chamber. The plateout fraction will be determined by subtracting the amount of material that reaches the filters.

Since the facility can operate with very high-burnup specimens of reasonable size, it is hoped that the fission-product concentration effect will be readily indicated.

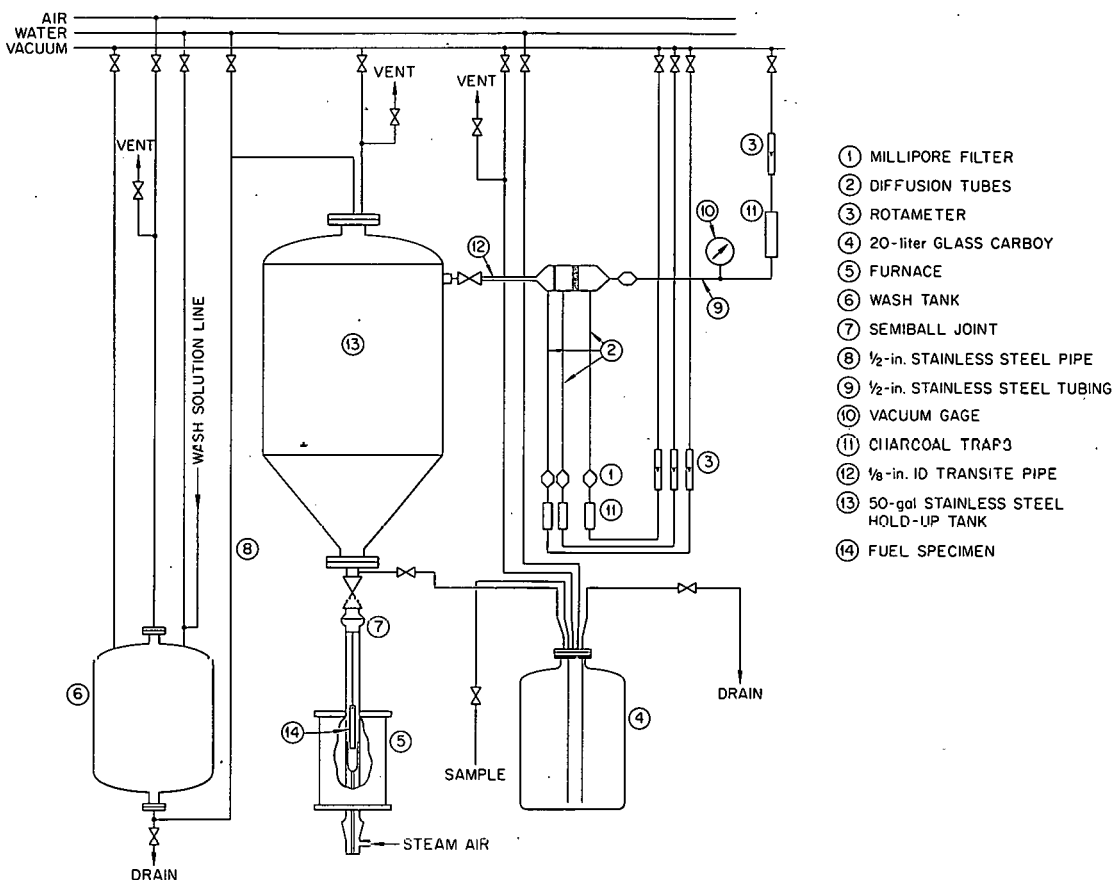


Fig. 7.1. Hot-Cell Containment Mockup Facility.

RETENTION OF RADIOIODINE IN A MOCKUP CONTAINMENT FILTER UNIT

As an integral part of the program to provide facilities for handling larger quantities of irradiated fuel materials, a test filter unit has been incorporated in the hot-cell containment mockup equipment shown in Fig. 7.2. The design is consistent with the concept that a model containment system should use filters and charcoal adsorbers in series to trap molecular as well as particulate activity. However, under some conditions extremely small particles may be formed which could penetrate the "absolute" filters presently provided.

Since no prior information existed on the size and concentration of particles resulting from melting uranium-aluminum alloy fuels, it was necessary to demonstrate the reliability of the containment filter system by a meltdown of a typical fuel plate

with a fission-product concentration likely to be encountered in an operating reactor. Other factors likely to influence the properties of the aerosol produced include the type of condensation nuclei in the atmosphere, the length of time available in the containment for agglomeration, and the nature of the piping through which the aerosol passes before reaching the filter unit. In the test, all these factors have been given some consideration.

The tests completed were conducted with bench-size equipment made of quartz and glassware in preparation for a large meltdown in the hot-cell equipment. These experiments were designed to determine the iodine retention efficiency of the materials and the arrangement proposed for use in the containment system of the Oak Ridge Research Reactor. Two runs were made under expected ORR air flow conditions, and the results obtained characterized two lots of commercial-grade coconut

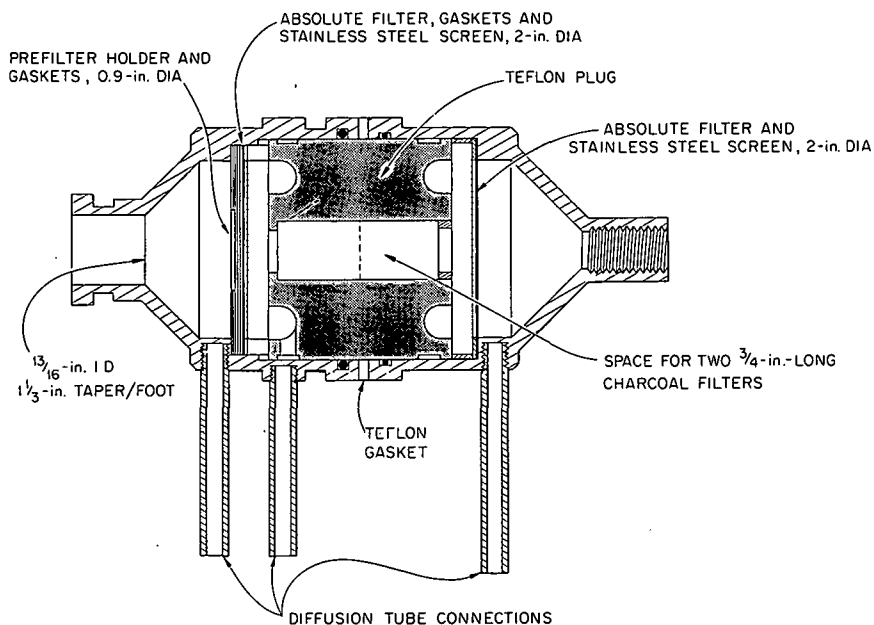


Fig. 7.2. Mockup of ORR Containment Filter and Charcoal System.

Table 7.2. Filter Tests of Radioiodine and Particulates Released by Melting Uranium-Aluminum Alloy in Air (Uranium-Aluminum Burnup Level 23.6%; Sample, ~1 g)

Temperature, 870°C for No. 6249 and 840°C for No. 45

Time at temperature, 60 min

Air flow, 3000 cc/min

	Charcoal No. 6249 ^a		Charcoal No. 45 ^b	
	Iodine (%)	Gross Gamma	Iodine (%)	Gross Gamma
Percent activity on filters ^c				
Roughing (95% aerosolve)	1.98	44.6	1.21	17.6
1st absolute (CWS-6) filter		1.3	0.887	1.7
1st charcoal (3/4-in. bed)	97.89	53.8	97.879	80.7
2d charcoal (3/4-in. bed)	0.113	0.1	0.015	0.02
2d absolute (CWS-6) filter		0.2	0.004	0
Hot charcoal (backup)	0.009	0.009 ^d	0.0013	0.002 ^d
Charcoal ratio, $\frac{\text{1st charcoal}}{\text{2d charcoal}}$	867	538	6520	4035
Efficiency of 1st charcoal filter	99.885	99.42	99.98	99.97
Efficiency overall filter	99.991	99.991	99.9987	99.998

^a4-20 mesh.

^b4-20 mesh, slightly finer than No. 6249.

^cPercent of activity reaching filters; gross gamma.

^dRepresents loss through the containment.

charcoal. The results were quite satisfactory, as shown in Table 7.2, although there is some reason to believe that the performance of a single charcoal bed in test No. 6249 may have been impaired by a slight atmospheric leak around the front cartridge. Otherwise the results suggest the presence of much more than 99% of the iodine in the molecular state under the conditions employed in the tests. The conclusion that the efficiency of the containment system is equally high whether tested with tracer iodine or with fuel-plate-released iodine seems to be warranted. The overall efficiency of the two beds in series in each case was greater than 99.99%.

IDENTIFICATION OF CHEMICAL SPECIES

Additional experiments with the sealed-tube iodine equilibration apparatus described in the

previous report¹ gave low values for the heat of evaporation of iodine for reasons that are not, at present, understood. A capsule containing 0.20 mg of iodine labeled with I¹³¹ gave a value of 12.0 kcal/mole, while a value of 11.1 kcal/mole was obtained with a similar capsule containing 5.1 mg of iodine. No gross contamination was observable in either capsule, but it is possible that traces of moisture, stopcock grease, or other impurities may have caused the deviation from the accepted value for the heat of vaporization of solid iodine (14.9 kcal/mole). Because of the difficulties experienced in developing a satisfactory method of identifying chemical species transported by gas streams, this line of endeavor is being discontinued, for the present, in order to undertake the studies of high-temperature iodine trapping methods described below.

¹C. J. Barton, M. E. Davis, and G. W. Parker, *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, p 41.

8. Iodine Removal from Oxidizing Gases at High Temperatures

C. J. Barton

G. W. Parker

M. E. Davis

REMOVAL OF RADIOIODINE FROM AIR BY EXCHANGE WITH INACTIVE POTASSIUM IODIDE AND BY PLATINIZED ALUMINUM OXIDE

Studies conducted at this laboratory and elsewhere have demonstrated that charcoal is a very efficient trap for molecular iodine. It is so efficient, in fact, that when a large amount of airborne iodine and other fission products liberated in a reactor accident are passed through a charcoal bed they will stop quite close to the front edge of the bed and the fission-product-decay heat could raise this part of the bed to temperatures above 300°C. Additional heat may be transferred from the overheated reactor core to the bed by the contaminated

coolant gas. Since charcoal will burn when exposed to air at high temperatures, it would be desirable to have alternate extraction methods that would be less susceptible to destruction under these conditions. Tests were performed with two different approaches to this problem during this period.

Preliminary experiments were conducted to test the effectiveness of iodide salts as a chemical-exchange medium for gaseous radioiodine and of activated aluminum oxide containing about 0.5% Pt (a hydroforming catalyst).¹ Concerning the iodide salts, of primary interest was (1) the ability of an

¹The authors are indebted to P. H. Emmett for suggesting the platinized alumina as a possible iodine extractant.

iodide to sorb or exchange with molecular iodine released in a gas stream and (2) the stability of the iodide in the presence of flowing helium and air at temperatures approaching the melting point of the salt. Related previous investigations were those of Fialkov and Nazarenko² who studied exchange reactions of iodine in systems with inorganic iodides but under quite different conditions than those employed in the experiments reported here. In the present tests, potassium iodide was used because it was available in granular form and it is one of the higher-melting iodides (mp, 686°C). A 3-in. bed of 10- to 16-mesh potassium iodide was prepared in a $\frac{3}{4}$ -in.-diam quartz tube. The bed was heated to 600°C by a tube furnace while air or helium flowed through it at different rates. The qualitative observations shown in Table 8.1 indicate that the bed was stable at least for several hours at 600°C in both helium and air when the gas flow rate through the bed was below about 10 linear feet per minute (lfm). At higher gas flow rates of both air and helium (23 and 46 lfm respectively) slight decomposition of the bed was noted in a few minutes. Instability of the bed was indicated by the presence of a thin deposit of potassium iodide on a cooler portion of the tube downstream from the bed and by the appearance of I₂ in a carbon tetrachloride trap through which the effluent gas passed. The house supply

²Y. A. Fialkov and Y. P. Nazarenko, *Ukr. Khim. Zh.* 19, 356 (1953).

Table 8.1. Stability of Potassium Iodide in Gas Streams at 600°C

Sweep Gas	Flow Rate (lfm) ^a	Observation
Helium	2.3 to 3.4	Only very slight visual indication of instability in 3 to 5 hr
Helium	46	Some apparent decomposition of iodide salt in 3 to 5 min
Air	6.7	Only very slight visual indication of instability in 3 to 5 hr
Air	23	Some apparent decomposition of iodide salt in 3 to 5 min

^aAir flow measured at room temperature.

of helium used in this experiment is known to contain both moisture and oxygen impurities.

Approximately 1 mg of iodine (I₂) tagged with 0.1 mc of I¹³¹ was released into a helium stream which flowed through a freshly prepared bed of potassium iodide at a rate of 10 lfm to observe the interchange of gas-borne molecular iodine with inactive I¹²⁷ in the potassium iodide bed which was heated to 500°C. The activity in the iodide bed was measured by means of a sodium iodide gamma-ray detector, and iodine that passed through this bed was caught on a charcoal trap which was monitored by a second sodium iodide detector. The tube was cooled periodically and removed from the furnace, and the distribution of I¹³¹ on the bed was determined by use of a $\frac{1}{8}$ -in. slit collimator and a gamma-ray detector. The potassium iodide was subjected to the following sequence of treatments: flowing helium at 500°C for 6.5 hr and at 600°C for 3.5 hr; flowing air at 500°C for 7.6 hr and at 600°C for 4.0 hr. In all cases the gas flow velocity was 10 lfm. Until the last treatment (air at 600°C), there was only slight evidence of decomposition of the bed, and no I¹³¹ activity could be detected in the charcoal trap at the discharge end of the tube. About 13 min after beginning the exposure of the bed to air at 600°C, more rapid decomposition of the bed was apparent, and also, over a longer period, a slow buildup of activity in the charcoal trap was noted. However, the activity in this trap had increased by only about 15% over the background of 2000 counts/min after 4 hr of exposure. A plot of I¹³¹ activity vs distance from the front edge of the potassium iodide bed after the first exposure to helium at 500°C for 6.5 hr and at the end of the sequence of treatments is shown in Fig. 8.1. The activity of I¹³¹ was never significantly above background at the exit end of the bed (3-in. position).

From these preliminary experiments, it appears that the effectiveness of a potassium iodide exchange bed is not greatly impaired by exposure to air at temperatures below 600°C. Even under conditions that resulted in some decomposition of the iodide bed, very little of the I¹³¹ escaped. It appears, therefore, that the chemical-exchange method holds some promise for the removal of gas-borne fission products and that further studies of this type are warranted.

Only a cursory examination has been conducted to date to test the effectiveness of platinized

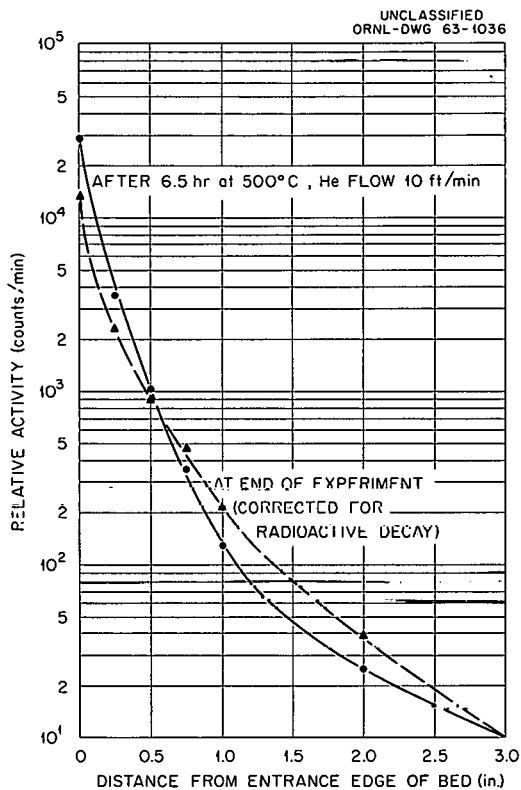


Fig. 8.1. Distribution of Radioiodine on Potassium Iodide Bed.

alumina as an iodine extractant. However, it appears that the platinized alumina is indeed a very effective adsorbent and can be expected to operate as both a heat sink and an iodine absorbent that might be effective for several hours at temperatures as high as 525°C. In Fig. 8.2, the distribution profile for radioiodine is shown for 30-mesh platinized Al_2O_3 (Girdler G-43) which, after loading with tracer I^{131} at 525°C, was heated 1 hr at each temperature (325, 425, 525, and 625) consecutively, while air was drawn through the bed at a rate of 600 cc/min. The resulting distribution on the front of the bed is shown. At 525°C, a small loss from a 3-cm bed depth resulted, while at 625°C almost 100% elution occurred.

It was observed that unplatinized Al_2O_3 was significantly less effective as an adsorbent than

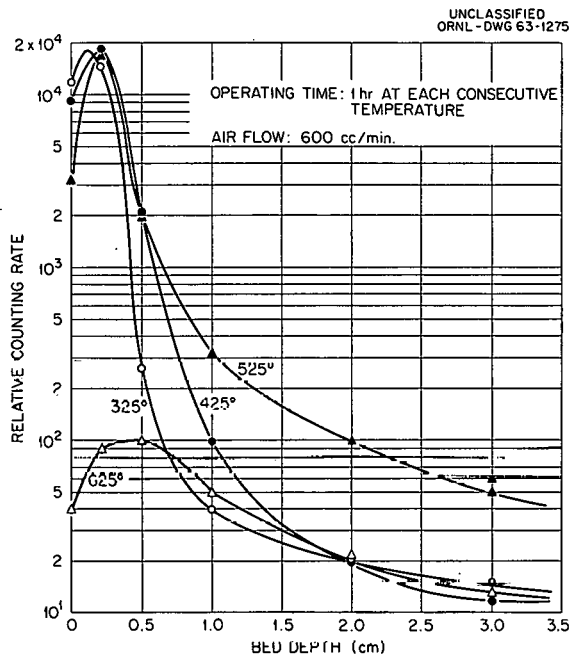


Fig. 8.2. Temperature vs Distribution Curve for Radioiodine on 0.5% Pt- Al_2O_3 Bed, 30 Mesh.

the platinized material, as were Linde molecular sieve No. WA 500 and refrigeration-grade silica gel. In the British Atomic Energy Program, the overheating problem is said³ to be combated by incorporating a large heat sink in the form of granite chips in the containment system. Ordinary gas coke is reported⁴ to have a lower adsorbent capacity for iodine than charcoal, but it is less combustible and may be examined later.

A more direct comparison of the high-temperature iodine retention properties of KI and platinized alumina is planned for the near future.

³C. M. Nicholls, private communication to G. W. Parker, June 1963.

⁴J. B. Morris and C. H. Rumary, *Tests on Absorbents for Iodine at Low Concentrations*, UKAEA Report AERE-R-4219, February 1963.

9. Attenuation of Fission-Product Escape by Intrinsic Processes

Certain processes which tend to limit the escape of fission products have been given more attention than others in assessing the potential hazard of reactor accidents. Credit is claimed for the partial retention of fission products by the fuel material during an accident. In various reactor designs, different amounts of credit are claimed for containment vessel performance and for engineered safeguards such as filtered ventilation systems. It is generally conceded that there may be significant attenuation of fission-product release by other processes which would occur naturally. Advantages of these processes have not been fully utilized in reducing the assumed consequences of reactor accidents for various reasons including complexity of the theoretical treatment of some of the processes, inability to describe satisfactorily the conditions under which these attenuation processes might take place, and uncertainty as to the possible magnitude of attenuation effects. Theoretical analyses of some of these attenuation processes will make it possible to judge whether the effects are of sufficient magnitude to warrant investigation in further detail and may suggest ways in which design changes can result in significant reduction of the potential hazard of reactor accidents. Two mechanisms which are of possible importance are diffusional deposition within a leak in a containment vessel, and agglomeration of particles during aging prior to filtration. Therefore, theoretical treatments were carried out to determine the reduction of fission-product release which would occur under various conditions as a result of these mechanisms.

DECONTAMINATION FACTORS FOR PARTICLE DEPOSITION IN CONTAINMENT VESSEL LEAKS

W. E. Browning, Jr.

R. D. Ackley

Very fine airborne particulate matter would probably constitute the form of a significant fraction of the radioactivity released from the reactor core in

the event of a major nuclear-reactor accident.¹ Consequently, it would be of considerable value when establishing criteria for the leak testing of containment vessels to have some idea of the expected extent of removal of fine particles, due to diffusional deposition, as the contaminated air escapes through undetected leaks which may occur as pinholes or cracks in the vessel wall. Diffusional deposition results from the transport by Brownian diffusion of particles to the boundaries of the leak where they are held by van der Waals forces.

For leaks where the smallest dimension is in the vicinity of 10μ , calculation indicates that the flow would be approximately viscous. Therefore, the equations of Poiseuille for cylindrical capillaries and that given by Knudsen and Katz² for laminar flow between infinite parallel planes were adopted for estimation of the air flows. These equations were combined, respectively, with equations for the fraction of particles penetrating cylindrical and rectangular channels to yield theoretical decontamination factors; for the cylindrical case, the equation of Gormley and Kennedy³ was employed and for the rectangular, that of DeMarcus.⁴ The resulting expressions for calculating the decontamination factors follow:

$$\text{cylindrical, } \log_{10} \text{D.F.} = 0.0867 + 203.2 \frac{\eta DL^2}{\Delta P d^4}; \quad (1)$$

$$\text{rectangular, } \log_{10} \text{D.F.} = 0.0386 + 39.35 \frac{\eta DL^2}{\Delta P b^4}; \quad (2)$$

¹W. E. Browning, Jr., et al., *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, pp 27-35.

²J. G. Knudsen and D. L. Katz, *Fluid Dynamics and Heat Transfer*, p 100, McGraw-Hill, New York, 1958.

³P. G. Gormley and M. Kennedy, *Proc. Roy. Irish Acad. Sec. A* 52, 163-69 (1949).

⁴Wendell DeMarcus and J. W. Thomas, *Theory of a Diffusion Battery*, ORNL-1413 (Oct. 16, 1952).

where

D.F. = decontamination factor or the ratio of the number of particles entering the channel to the number penetrating,

η = coefficient of viscosity of air, poises,

D = diffusion coefficient of particle in air, cm^2/sec ,

L = length of leak or wall thickness of containment vessel, cm,

ΔP = pressure drop through leak, dynes/ cm^2 ,

d = diameter of cylindrical leak, cm,

b = smallest dimension of rectangular leak or, for the model, the distance between parallel infinite planes, cm.

Strictly speaking, ΔP should be infinitesimal in applying Eqs. (1) and (2); however, for convenience in making the calculations for the results to be

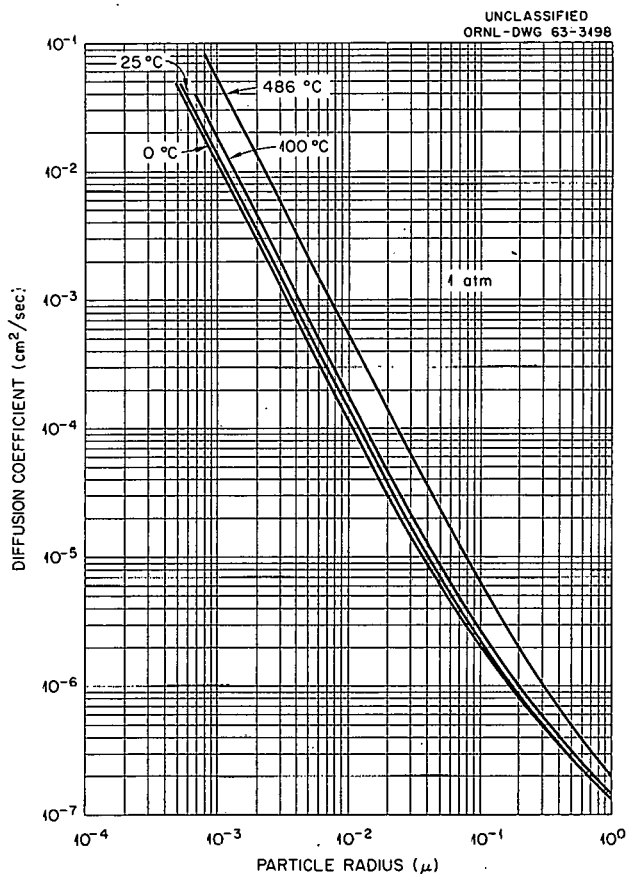


Fig. 9.1. Calculated Diffusion Coefficients for Particles in Air.

presented, this restriction has been ignored. The particle diffusion coefficients employed were obtained from the 25°C curve of Fig. 9.1. The calculations for the diffusion coefficients in the figure were performed using the Einstein-Stokes equation with the Cunningham slip correction as evaluated by Langmuir from the determinations of Millikan and others but with the reevaluated constants given by Thomas.⁵ Curves for the other temperatures are rather incidental in the present connection but are included because they were already available and may be of some interest.

Decontamination factors for a particular set of conditions are shown for cylindrical and rectangular leaks in Figs. 9.2 and 9.3 respectively. For these conditions, and depending on whether the geometry is rectangular or cylindrical, leaks as small as 5 or 10 μ in nominal size would apparently permit the escape of an appreciable amount of the radioactivity associated with particles $\sim 1 \mu$ in diameter. For 0.3- μ particles, the degree of decontamination would be expected to be high only if there are no leaks larger than, say, 5 μ . However, in the case of 0.05- μ particles, good decontamination should result if leaks larger than 10 μ are absent. These comments, which apply to the conditions corresponding to the figures, indicate the desirability of estimating particle-size distribution vs time data for the radioactive aerosols produced by a nuclear accident.

A brief note regarding leak detection may be in order here. If a leak is observed, for the Fig. 9.2 or 9.3 conditions, that forms a 1-mm soap bubble per second, calculation indicates a cylindrical hole with a diameter of about 20 μ or a 2- μ crack for an assumed width of 1 cm. Thus, the leak dimension range under discussion, 5 to 30 μ , may correspond roughly to the lower limit for practical leak detection. Containment vessel design should evidently provide for good accessibility for leak testing, particularly in the case of suspect areas such as welds; also, further development of leak-detection techniques may be warranted.

In considering diffusional deposition of particles for other wall thicknesses or different pressure drops, the corresponding decontamination factors can be estimated using the figures along with the application of an appropriate adjustment based on

⁵J. W. Thomas, *The Diffusion Battery Method for Aerosol Particle Size Determination*, ORNL-1648 (Dec. 14, 1953).

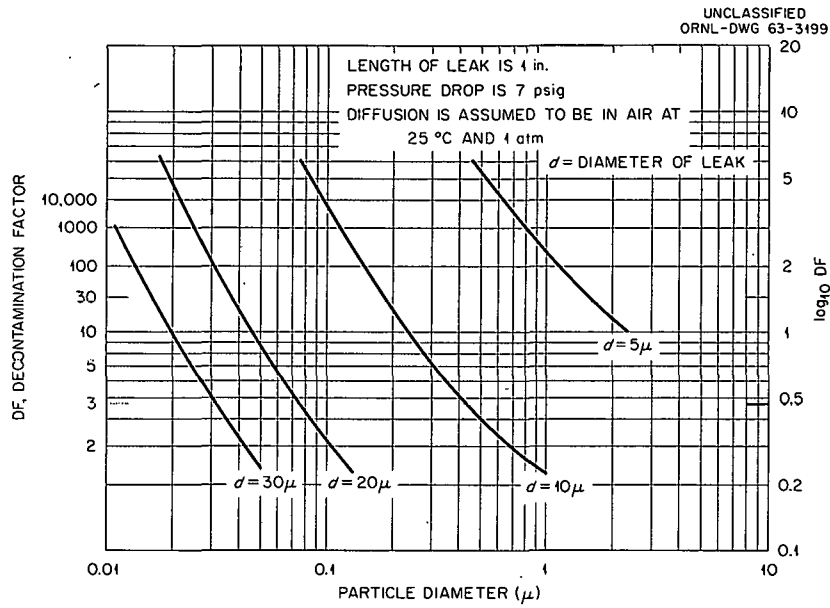


Fig. 9.2. Decontamination Factors Resulting from Diffusional Deposition of Particles in Cylindrical Leaks in Reactor Containment Vessels.

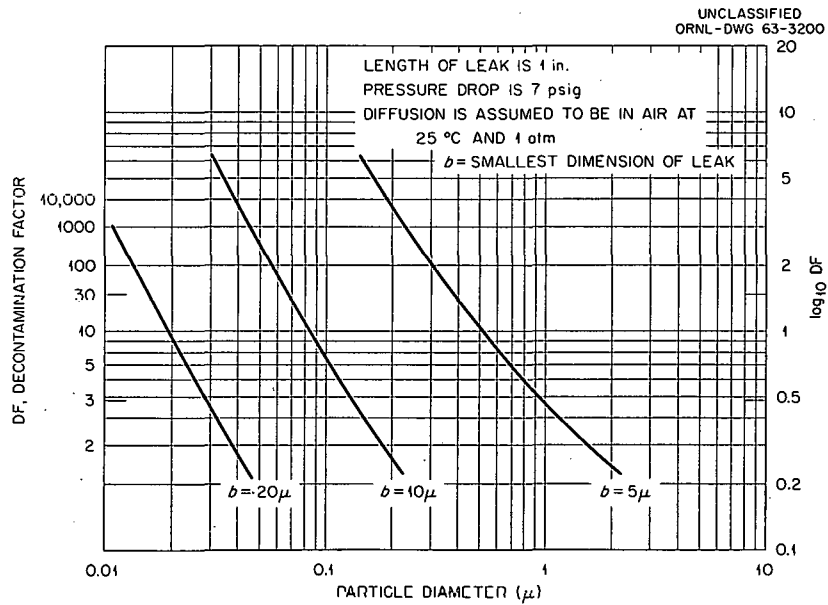


Fig. 9.3. Decontamination Factors Resulting from Diffusional Deposition of Particles in Rectangular Leaks in Reactor Containment Vessels.

Eqs. (1) and (2). Alternately, for some given situation, direct use of Eqs. (1) and (2) together with Fig. 9.1 might be preferable.

EFFECT OF PARTICLE AGGLOMERATION ON THE PENETRATION OF FILTERS IN DOUBLE-CONTAINMENT SYSTEMS

W. E. Browning, Jr.

M. H. Fontana

The penetration of filters by fission products adsorbed on submicron particulate matter has been of some concern to those responsible for the performance of nuclear-reactor containment systems employing filtration as a means of limiting activity release. The efficiency of absolute particulate filters is specified to be >99.95% for 0.3- μ -diam particles, but the filtration efficiency is not known, with comparable certainty, for particles of substantially smaller size, for example, in the range 20 to 40 A. It is known, however, that particulate matter of a high particle-count density per unit volume which would exist after a reactor accident would agglomerate at a rapid rate to a smaller number of larger particles.

A simplified analysis was performed of the effects of particle agglomeration on the activity release from a containment system that depends on particulate filtration for its effectiveness. The case treated here is that of a dispersion of activity into a primary volume, such as the first containment vessel, and its leakage into a secondary volume in which a sweep air flow is maintained and filtered before it is exhausted into the surroundings. The secondary barrier is thus made effective in that all leakage from the surroundings flows into it and all activity that is released is filtered. Some examples of this kind of containment are the NS "Savannah" and the Hallam reactor.

A calculation of particulate agglomeration was performed on a simplified model, which was shown to be conservative. Since iodine is of major concern in filtration problems, it was chosen to illustrate the particle behavior. Only the particulate form of iodine was treated. It was assumed that iodine remaining in vapor form was removed by adsorbers. The analysis was performed in the following manner: (1) It was assumed that all the iodine isotopes existing in the reactor core after

full-power, full-life operation were released at the time of the accident and evenly dispersed within the primary containment vessel as an infinite number of infinitely small particles; (2) the iodine agglomerated as a monodisperse pseudoaerosol of pure iodine yielding particle sizes as a function of time; (3) all the aerosol with a particle size <0.3 μ that leaked out of the primary containment penetrated the secondary containment filtration system; and (4) the amount of iodine being released during this time period of zero filter efficiency was calculated.

The agglomeration calculations were performed using the relation:⁶

$$\frac{1}{n} = \frac{1}{n_0} + Kt, \quad (1)$$

where

- n = number of particles per cm^3 at time t , cm^{-3} ,
- n_0 = number of particles per cm^3 initially, cm^{-3} ,
- t = time, sec,
- K = agglomeration coefficient ($\text{cm}^3/\text{particle-sec}$) taken as 5×10^{-10} (ref 6).

The value n was related to the concentration of matter in air by the equation

$$\frac{m}{V_1} = \rho \frac{\pi}{6} d^3 n, \quad (2)$$

where

- m = total mass of material making the particles, g,
- V_1 = total free volume of the primary containment, cm^3 ,
- ρ = density of the particle, g/cm^3 ,
- d = diameter of particle, cm.

Substituting Eq. (2) into Eq. (1) yields the time required to reach a given diameter,

$$t = \frac{d^3 \pi \rho}{6(m/V_1)K} - \frac{1}{n_0 K}. \quad (3)$$

Since n_0 is large, the last term is negligible.

⁶H. L. Green and W. R. Lane, *Particulate Clouds, Dusts, Smokes, and Mists*, pp 126 et seq., E. and F. N. Spon, Ltd., London, 1957.

The diluting effect of the sweep gas in the secondary containment was considered because the filters were located in this system. The ratio of the rate of activity released from the secondary containment to the activity release rate from the primary containment can be shown to be:

$$\frac{C_2 Q_{23}}{C_1 Q_{12}} = 1 - e^{-(Q_{23}/V_2)t}, \quad (4)$$

where

C_2 = concentration of material of interest in the secondary containment, curies/cm³,

Q_{23} = sweep gas volumetric flow rate, cm³/sec,

C_1 = concentration of activity in primary containment, curies/cm³,

Q_{12} = volumetric leak rate from the primary containment, cm³/sec,

V_2 = volume of secondary containment, cm³.

The integrated release from the secondary containment up to time t compared with the rate of release from the primary containment can be shown as

$$\frac{A_{23}}{IR} = t - \frac{V_2}{Q_{23}} [1 - e^{-(Q_{23}/V_2)t}], \quad (5)$$

where

A_{23} = total integrated activity released to time t , curies,

I = total activity inventory in the primary containment available for release, curies,

R = primary containment release rate, %/sec.

Figure 9.4 is based on Eq. (3) and shows the particle size as a function of time for six values of (m_1/V_1) and a particle density of 4.93 g/cm³, corresponding to elemental iodine. Curve E shows the values for the NS "Savannah," selected as an example of a containment system.

Figure 9.5 shows the performance of the secondary containment sweep gas system, relating the total activity released up to time t for three values of the volume change times (V_2/Q_{23}) . Curve E of Fig. 9.4 is superimposed on Fig. 9.5 to show the procedure for finding the worst-case operation of the system, assuming that all activity, I , is in particulate form and all that is released from

the primary containment prior to the time that the critical particle size is attained passes through the filtering system in the secondary containment. For example, if 0.3 μ is chosen as the critical particle diameter, it can be seen that if the core inventory of iodine is the only material available to make particles, this size will be reached in 235 sec. The NS "Savannah" has a secondary containment volume change time of 900 sec, and this curve shows that the total release to the atmosphere is $A_{23}/I = 26R$. Since the NS "Savannah" primary containment has a maximum leakage rate of 3%/day,

$$R = \frac{0.03}{8.65 \times 10^4} \text{ sec}^{-1} = 3.47 \times 10^{-7} \text{ sec}^{-1}.$$

The ratio of total activity released to that available is

$$\frac{A_{23}}{I} = 26(3.47 \times 10^{-7}) = 9.02 \times 10^{-6}.$$

Table 9.1 shows the core inventory and amounts released to the surroundings for the NS "Savannah" for the case of 0.3- μ critical aerosol diameter.

This analysis of the effect of agglomeration is based upon established principles or upon assumptions which are clearly pessimistic. Equation (1) has been shown to hold ever since reliable methods of counting the particles in smoke clouds have been available.⁶ Theoretical expressions for coagulation of monodisperse particles can be shown to be:

$$\frac{1}{n} - \frac{1}{n_0} = \frac{4RT}{3\eta N} \left(1 + \frac{Al}{r}\right)t, \quad (6)$$

where

R = gas constant,

T = absolute temperature,

η = viscosity of the medium,

N = Avagadro's number.

The term $[1 + (Al/r)]$ is the Cunningham correction for non-Stokes behavior of very small particles where A is a constant (~ 0.9), l is the mean free path of the molecule, and r is the radius of the particle. From Eq. (6) it can be seen that the

coagulation coefficient

$$K = \frac{4RT}{3\eta N} \left(1 + \frac{AI}{r} \right)$$

would decrease as the radius increases. Experimentally determined graphs of t vs $1/n$ are straight lines within the limits of experimental error except for the case of very small particles, for which there is an indication of curvature. The use of this theoretical expression for K for air at standard conditions and a standard stearic acid cloud yields $K = 5.1 \times 10^{-10}$ cm³/sec. The use of $K = 5.00 \times 10^{-10}$ cm³/sec in this analysis in Eq. (1) appears justified on a conservative basis.

The artifice of assuming that all the iodine coagulates into particles of pure iodine is a useful concept that can be shown to be conservative. Actually, in the case of a real accident, some of the iodine would remain in the vapor form and some would deposit on particles caused by debris and smoke concurrent with fuel meltdown. Taking the case of all the iodine depositing

on particles of other matter, then the m of Eq. (3) must include the iodine plus the other debris and must be larger than in the case of pure iodine; for example,

$$m_t = m_0 + m_1, \quad (7)$$

where

m_t = total mass, g,

m_0 = mass of foreign matter, g,

m_1 = mass of iodine, g.

It can be seen from Eq. (3), however, that the time required to reach a critical, filterable diameter is directly proportional to ρ_i/m_t . Since $\rho_i/m_t = 1/V_p$, where V_p is the volume taken up by the particle, substitution into Eq. (3) shows that

$$t = \frac{d^3 \pi}{6K(V_p/V_1)}; \quad (8)$$

that is, the time to reach a given diameter is inversely proportional to the ratio of the volume

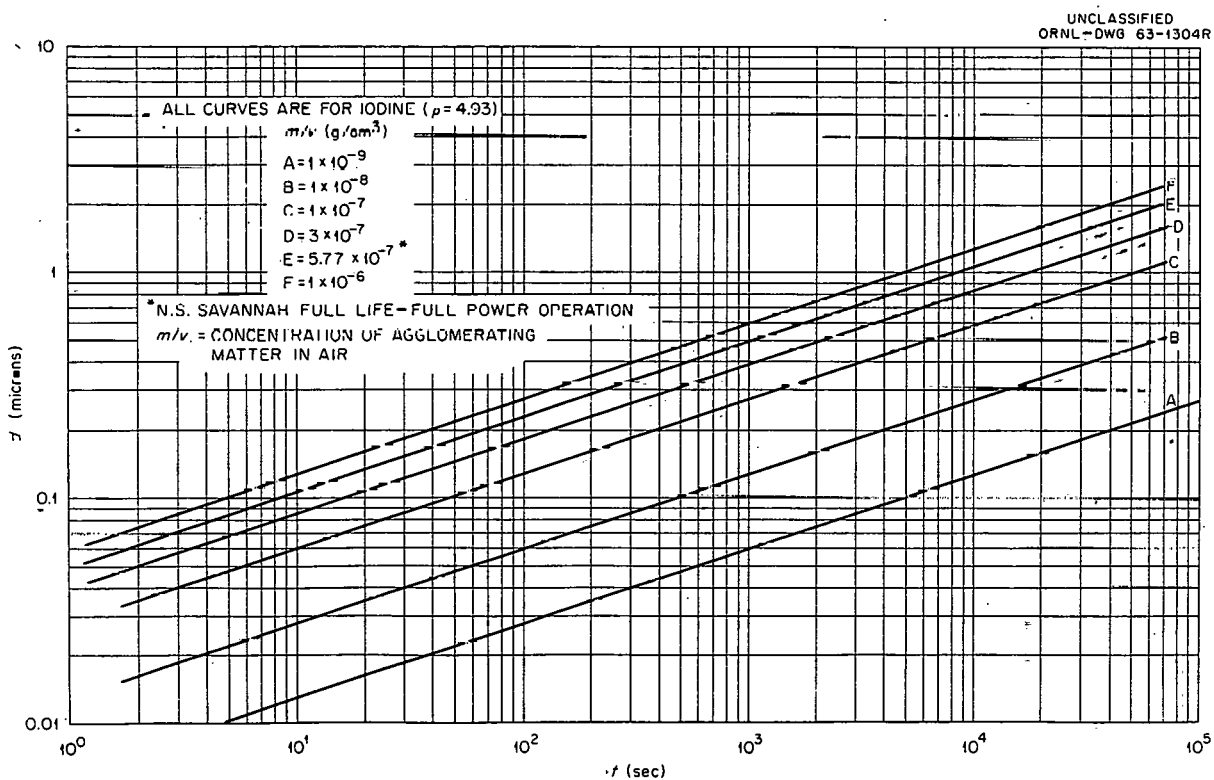


Fig. 9.4. Particle Size vs Time as Controlled by Agglomeration.

taken up by the particles to the total free volume of the containment vessel. [Note, however, that V_p/V_1 must be very small for Eq. (1) to hold.] Equation (8) shows that if foreign matter is added to the iodine, the time at which filtration is effective will be shorter.

The usual effect of a size distribution in a cloud of particles is to promote agglomeration at a faster rate than would occur in a cloud of equal-sized particles.⁶ It appears that the assumption that all particles of a size below 0.3μ penetrate the filter and the faster agglomeration rate of the polydisperse particles introduce a greater degree of conservatism to this worst-case analysis than could be disputed by neglecting the

trace amount of small particles that has not undergone agglomeration up to the time of filtration.

Table 9.1. Activity Release for the NS "Savannah" Conditions

Nuclide	Core Inventory ^a (curies)	Amounts Released (curies)
8.05-day I ¹³¹	2.29×10^4	0.206
20.8-hr I ¹³³	2.05×10^6	18.5
6.68-hr I ¹³⁵	2.01×10^7	181

^a600 days at 69 Mw.

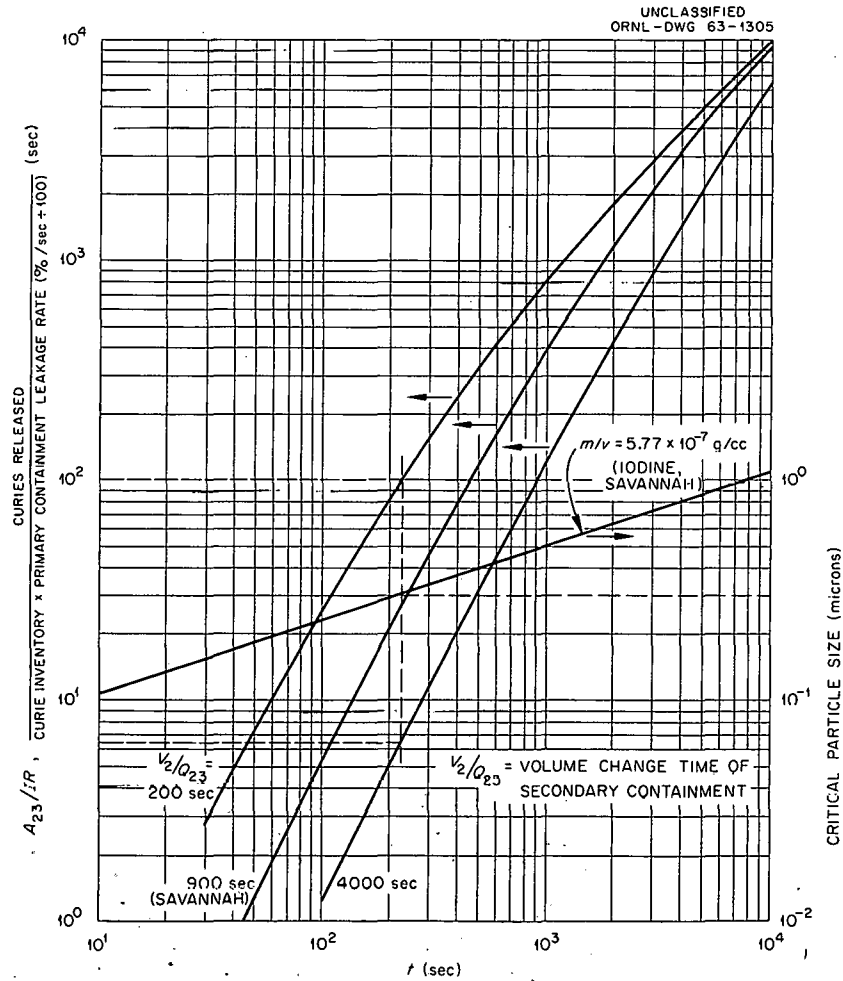


Fig. 9.5. Total Activity Released During Period of Zero Filter Efficiency per Release Rate vs Time.

From the results of this simplified analysis, it appears that the extent of fission-product penetration of filtration units by transport of particulate matter may be limited because with the high particle-count density which would exist after a catastrophic reactor accident, the particles would agglomerate in a very short time to a size readily

removable by filters. This analysis shows that agglomeration occurs at a rate fast enough to prevent significant penetration of the filtration systems even if all particles of a size smaller than 0.3μ were assumed to penetrate the filters. In one example, the calculated amount of iodine released would be less than 10^{-5} of that available.

Part III

Containment Engineering

Reactor Division

S. E. Beall, Division Director
W. B. Cottrell, Coordinator

10. Nuclear Safety Pilot Plant

L. F. Parsly

P. P. Holz

The purpose and design of the Nuclear Safety Pilot Plant were described in the previous semi-annual report.¹ Work during the past six months has been principally in design, installation, fabrication and procurement of components, and installation of equipment and piping in cell B, Building 7500. In addition, consideration has been given to the possible operating program for the facility, and operating procedures have been developed as a necessary step before completing design details.

STATUS OF DESIGN AND CONSTRUCTION OF NSPP

There have been no significant changes in the design concept during this period. The design effort has been concentrated on the preparation of drawings for installation of equipment and piping.

¹M. H. Fontana and C. G. Lawson, *Nuclear Safety Program Semiann. Progr. Rept. June 30, 1962*, ORNL-3319, pp 51-62.

On the process system proper, 38 drawings were issued and 18 drawings were revised during the report period.

Installation of equipment and piping was started in April and is approximately 60% complete. All components except the gas sampling train and the furnace have been set in place. Piping of the air sampler system, the decontamination system, and the steam system in the cell is essentially complete. Figures 10.1-10.3 show the status of the installation in cell B at the end of the report period.

The instrument flowsheets and panelboard layout have been approved. Procurement of process instruments, including control valves, is essentially complete. Fabrication of the control panel is under way. Electrical design is complete.

Rough-draft operating procedures have been prepared, although final procedures cannot be established until the plant is complete and the remote operations can be rehearsed in place. A hazard report for the facility has also been drafted.

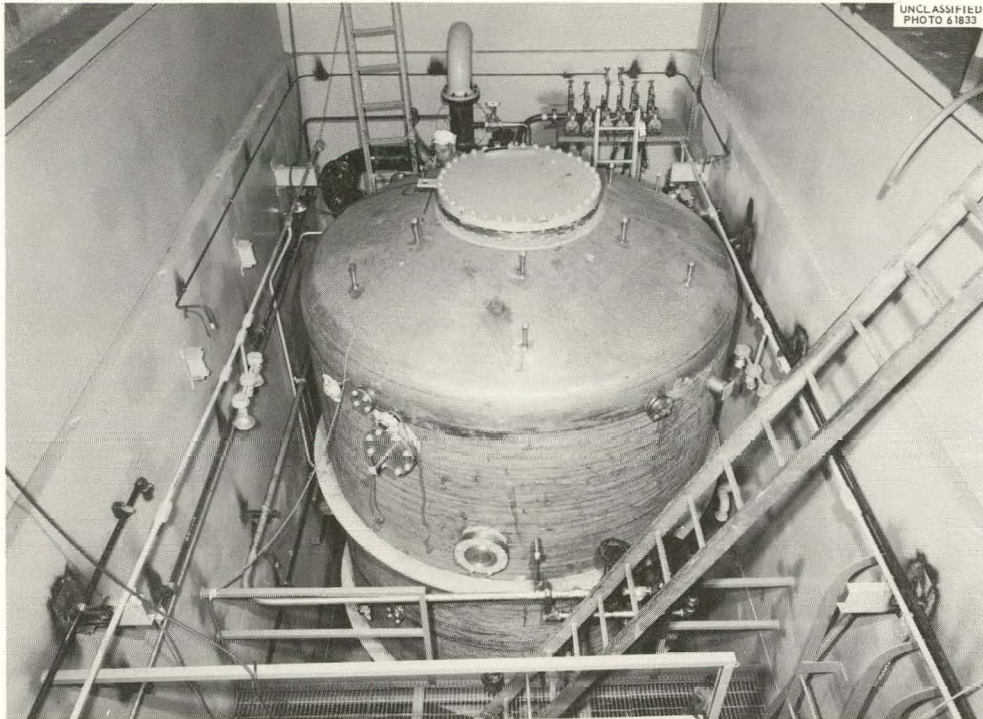


Fig. 10.1. View of Installation in Cell B from South End.



Fig. 10.2. South End of Cell B, Showing Piping Penetrations.

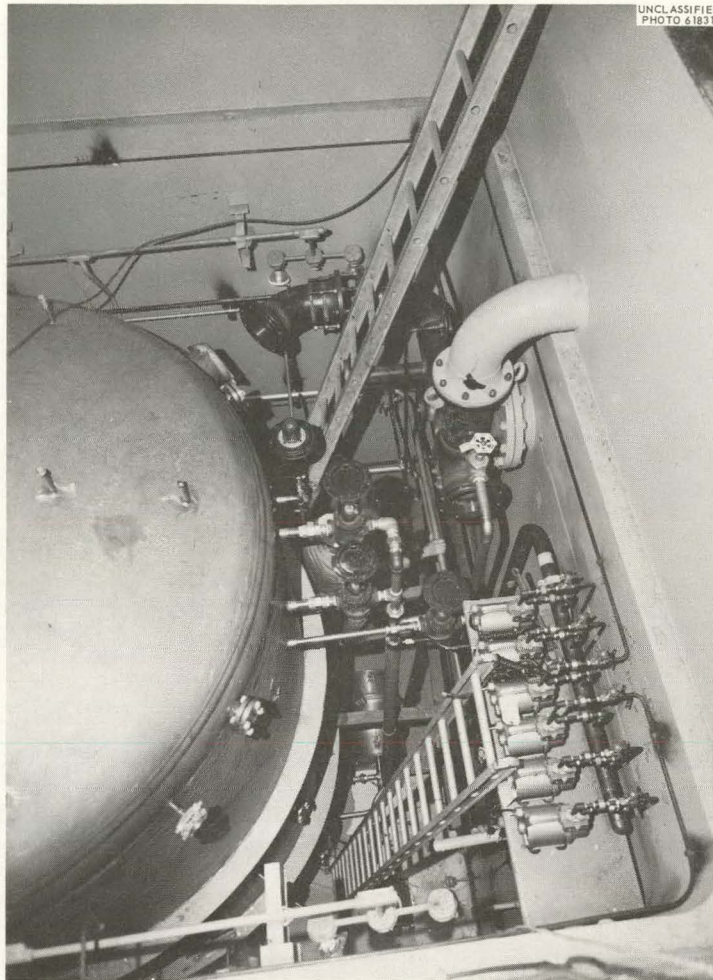


Fig. 10.3. Equipment in North End of Cell B.

REMOTE MAINTENANCE

Following a general review and a study of flow-sheets, equipment, equipment layout, and piping of the NSPP cell at Building 7500, a tentative operating procedure was prepared to establish tooling requirements for routine maintenance operations. In addition, the procedure served as a guide for establishing the carrier requirements for the transfer of various test-component equipment from the irradiation facility to the cell and, subsequently, from the cell to glove boxes, hot cells, analytical building, or burial ground.

Two drawings were issued to cover building modifications — a reversal of a bay crane and a

new shielding block arrangement. A jib boom was designed to provide additional hoisting capacity. Five additional drawings were certified for the procurement of a sectional seal pan to be placed above the lower cell shielding blocks. These pans will initially be used as bolted assemblies, including rubber seal strips. Their design, however, incorporates provisions for eventual seal welding, which may be required for high-radiation-level tests.

A total of eight drawings were released to show a variety of insert and attachment adaptations to existing 7500 area Vitro shielded carriers. Space up to 17 in. in diameter and 54 in. high is now available within the carriers for shielded transportation of large items.

Twenty-five tool drawings were issued for miscellaneous tool fabrication as follows:

1. remote tooling for Snap-Tite Disconnects, both for horizontal and vertical connector make and break operations;
2. remote tooling for fallout sample removal;
3. remote-controlled, torque-operable socket wrenches for both horizontal and vertical bolt manipulations;
4. a low-end offset attachment for the above socket wrenches;
5. self-locking socket attachments for in-cell storage;
6. remote-handle thermocouple disconnect tools;
7. remote-handle electrical disconnect tools;
8. furnace boat handling tools, both for pilot plant cell and hot cell use;
9. remote torque-controlled valve operators;
10. remote Grayloc Disconnect tooling;
11. remote utility grapples;
12. sheaths for remote viewing apparatus (omniscopes);
13. rod-handle-supported, 500-w caged lights;
14. miscellaneous remote handling tools – hooks, bails, rods, etc.

FURNACE DEVELOPMENT

An AVCO Corporation plasma-gun system with a PS 350 series 42-kw power source and control console was adapted to generate plasma capable of melting UO_2 .

A prototype furnace, built from pipe with an AVCO PG 100-1 gun mounted vertically and containing a screw-driven carrier within, completed the test facility for meltdown of nonirradiated EGCR-type UO_2 pellets. Considerably early, tests were run to check adaptability, feasibility, and reliability of such systems for use in detailed study of fission-product release from fuel-element configurations.

Design information showed that UO_2 pellets could be melted with the carrier moving under the torch at a speed of 1 in./min when the top of the specimen was mounted $2\frac{1}{2}$ in. below the torch and the current setting was 700 amp. Uranium dioxide temperatures could be maintained at lower levels by reducing the current.

It has been demonstrated that the melting may be observed by viewing the specimen through high-density optical filters normally used by welders. It has also been demonstrated that a Leeds and Northrop matching wire optical pyrometer can be used to measure the temperature of molten UO_2 in the presence of the plasma. Surface readings between 2700 and 2800°C were measured when the UO_2 began to melt while being heated with the plasma gun.

The final design of a furnace for the Building 7500 cell was based on the prototype experimental test results. The new furnace is fabricated from high-grade stainless steel materials employing controlled containment welding techniques. Design provisions also incorporate complete in-cell maintenance capability, including in-place charge loading and unloading, and in-place decontamination. The furnace shown in Fig. 10.4 essentially consists of a two-part construction, a trough bottom section with internal tracks for the carrier or "boat," and a stuffing box drive end connector as well as a rectangular lid which includes an attached tantalum heat shield, the torch, and off-gas line to the containment vessel pipe connectors, along with a pyrometry and a visual observation port. Both furnace parts and the "boat" are water jacketed. The furnace halves bolt together on a leak-detectable dual rubber O-ring-sealed joint. All components of the furnace, including a specially built torch and torch disconnect joint, are built to be remotely maintainable from an overhead maintenance shield. The "charge carrier boat" capable of handling fuel elements up to 2 in. in diameter and 8 in. long includes a primary containment seal lid cover attachment to permit contained transfer of charged boats from the loading cell to the furnace, and of the melted residue from the furnace to the analysis hot cell. A separate boat drive attachment is coupled to the furnace to permit remote changeout of either furnace or drive when necessary.

Thirty-two certified drawings were issued for the construction of the furnace and the torch and its disconnect. Shakedown tests are under way.

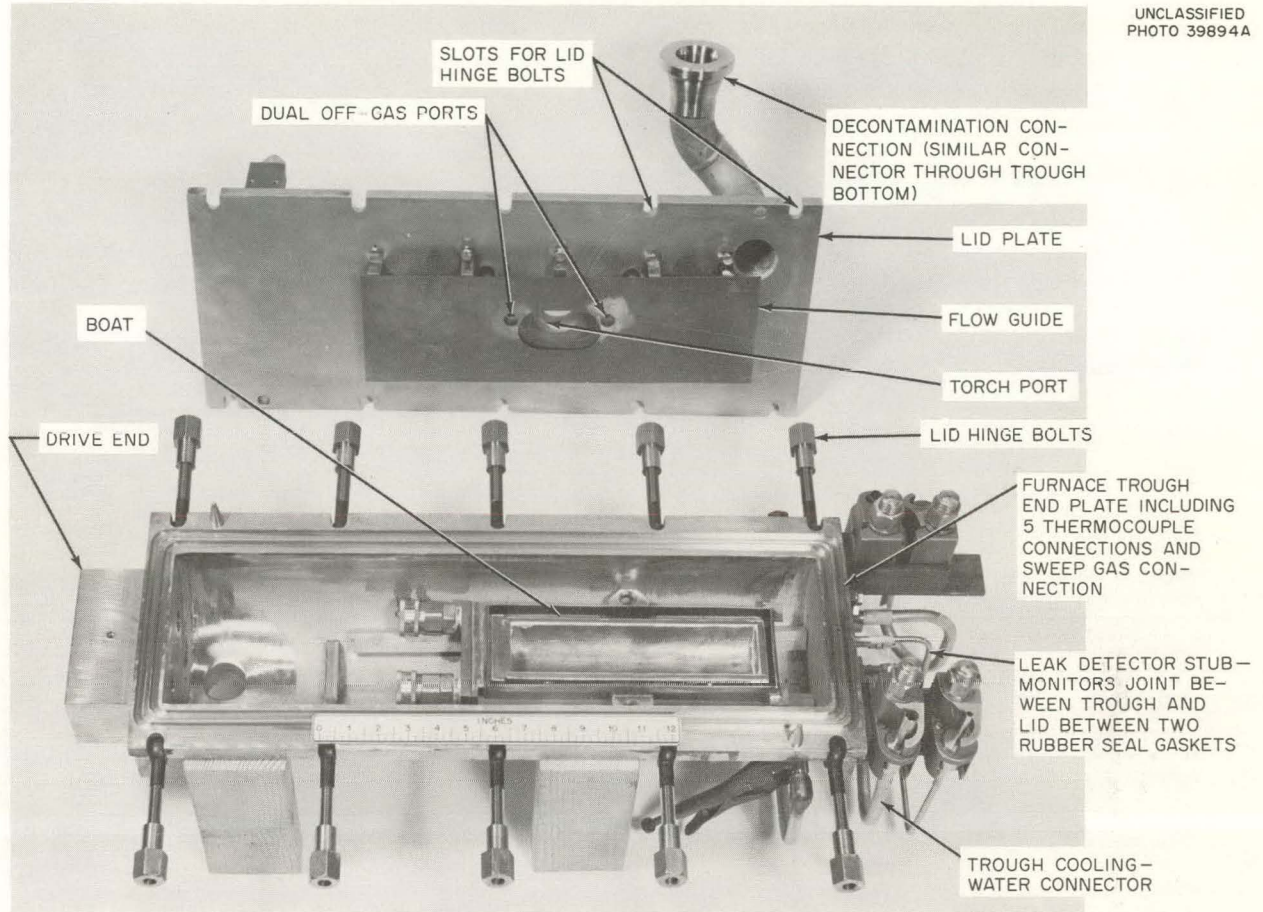


Fig. 10.4. Nuclear Safety Pilot Plant Furnace. Photo taken during fabrication.

11. Reactor Containment Handbook

T. H. Row

The purpose of the *Reactor Containment Handbook* is to provide information that will be useful in the design of reactor containment systems. The majority of the chapters are being prepared by personnel at ORNL, while four chapters are under subcontract to Bechtel Corporation. Completion of the handbook is scheduled for fall, 1963.

During the winter of 1963, J. R. Buchanan and H. B. Piper visited designers, constructors, and operators of several nuclear reactors to get information for Chapter 8 of the handbook, "Specific Containment Systems." Much of the information in the chapter, which deals with the application of specific types of containment systems to the

various reactors, was obtained from available literature. The visits were necessary, in part, to obtain information not available in the literature but more importantly to aid in understanding the implications of each item of information as it pertains to its associated containment concept. Sites visited by J. R. Buchanan and reactors discussed were as follows:

Company	Reactors Discussed
General Electric Co.	VBWR Humboldt Bay Dresden Big Rock Point PRTR
Bechtel Corporation	VBWR Dresden Humboldt Bay Big Rock Point Hallam
Pacific Gas and Electric Co.	Humboldt Bay
Atomics International	Hallam Piqua
Rural Cooperative Power Association	Elk River
Allis Chalmers Manufacturing Co.	Pathfinder
Sargent and Lundy	Elk River Containment concepts in general
Commonwealth Edison Co.	Dresden
Chicago Bridge and Iron Co.	Containers built by CB&I
Stone and Webster Engineering Corporation	Yankee CVTR Ravenswood PWR Conn. Yankee
Yankee Atomic Electric Co.	Yankee
Consolidated Edison Co.	Indian Point Ravenswood
Westinghouse Corporation	Yankee CVTR PWR Saxton Ravenswood

Sites visited by H. B. Piper and reactors discussed were as follows:

Company	Reactors Discussed
Babcock and Wilcox Co.	NS "Savannah" Indian Point
Carolinas-Virginia Nuclear Power Associates	CVTR
Savannah River	HWCTR

Preliminary drafts of the four chapters under sub-contract to Bechtel Corporation were submitted. These have been reviewed by ORNL staff members and comments forwarded to Bechtel for use in preparing the final draft, which is expected in October. Included in these four chapters are: Chapter 7, "Containment Proof Tests"; Chapter 9, "Design Details"; Chapter 10, "Containment Accessories"; and Chapter 11, "Economics."

Chapter 2, "Codes, Criteria, and Regulations," and Chapter 4, "Energy Sources," have been issued as internal memoranda and are currently being reviewed by ORNL personnel and external reviewers. Chapter 3, "Radioactivity - Generation and Transport," is completed and will soon be issued as an internal memorandum.

Chapter 5, "Analytical Techniques," is being written, with an expected completion date of September 1. In connection with Chapter 5, two sections have been prepared by firms participating in the AEC's Nuclear Safety Program. Stanford Research Institute has written a section on "Penetrability of Shells," while Armour Research Foundation has prepared a section on "Analysis of the Response of Containment Vessels to Internal Loading in the Range Approaching Failure."

Preparation of approximately 70% of the material for the handbook has been completed. Chapter 1, "Introduction," and Chapter 12, "Containment Research," will be completed after the other sections are in final form. All chapters will be reviewed by ORNL and selected external reviewers and will be updated before being issued in final form in late 1963.

An important part of the program will be the continued updating of the handbook in order that it will reflect the current and most valid design material available.

Part IV

Nuclear Safety Information Center

Reactor Division

S. E. Beall, Division Director
W. B. Cottrell, Coordinator

12. Nuclear Safety Information Center

W. B. Cottrell

J. R. Buchanan

A Nuclear Safety Information Center (NSIC) is being established at ORNL to serve as a focal point for the collection, storage, evaluation, and dissemination of nuclear safety information generated throughout the world. The Center will assist in coordinating the national effort in nuclear safety research and development and serve as a referral point for certain safety questions generated within the nuclear community. Accordingly, the Center will compile, index, and evaluate currently available information including experimental data, analytical studies, and actual experience relevant to nuclear safety technology and will make the information available to governmental agencies, research and educational institutions, and the nuclear industry. The Center will become operational about July 1.

SCOPE AND STAFF

The Center's activities are initially being limited to six specific areas of nuclear safety. A professional staff member, active in a designated area, will maintain continuous analytical and evaluation programs within the subject area. Approximately one-half man-year of effort will be spent in each of

the areas. The initial organization of the Center is as follows: W. B. Cottrell, Director; J. R. Buchanan, Assistant Director; and Juanita Perrou, Secretary.

The six specific areas of nuclear safety and the staff members in charge of the programs are listed below.

Containment of Nuclear Facilities (J. R. Buchanan). — This subject area will encompass all aspects of containment for reactors, radiochemical plants, hot cells, sources, etc., and will include such aspects as design considerations, leakage, penetrations, structural integrity, and testing.

Fission-Product Release, Transport, and Removal (G. W. Keilholtz). — The release of fission products from various materials and their movement within the containment system are included in this subject area. The release may be from any source by any mechanism, although the meltdown of a reactor core due to lack of adequate cooling is most frequently considered. The transport of the activity involves the physical and chemical characterization of the released radioactive material as well as the various mechanisms (i.e., deposition, adsorption, filtration, fallout, etc.) which would attenuate the concentration of the released activity within the containment system.

Meteorological Considerations (F. A. Gifford, USWB, and W. F. Hilsmeier, USWB). – Meteorological considerations are important to nuclear facilities not only for accident consequence evaluation but also for design consideration. In addition, the proposal of space vehicles has extended the height of interest of atmospheric dispersions from a few hundred feet above ground (where it is not completely understood) to a few hundred miles above ground (where we are only beginning to appreciate some of the processes involved).

Nuclear Instrumentation, Control, and Safety Systems (C. S. Walker). – The design of control and safety systems for various nuclear processes as well as instrumentation and hardware to effect the desired actions are included herewith. The specification of instrumentation, the concepts of coincidence, redundancy, and "fail-safe," the degree of reliability, the adequacy of shutdown margins, the design features of different mechanical devices, and related subjects are involved.

Radioactive Effluent Control, Monitoring, Movement, and Dosage (K. E. Cowser). – This subject area includes all aspects of the intentional or accidental release of activity to the environment except for the dispersion of airborne activity, which will be covered in the section entitled "Meteorological Considerations." Liquid waste and stack monitoring, movement of activity in soil and water, uptake of activity by flora and fauna, consideration of means by which the various processes may be enhanced, as well as the determination of their ultimate exposure of persons are included here.

Reactor Transients, Kinetics, and Stability (W. K. Ergen and P. H. Pitkanen). – This subject includes the various studies both analytical and experimental in which the transient behavior of reactors and criticality accidents are studied.

PHYSICAL PLANT

Arrangements have been made for several offices and necessary working space for the Center in the building (9711-1) occupied by the Y-12 Technical Library. These quarters will probably not be ready until December 1963, however. In the meanwhile the Center is occupying temporary quarters in a Library storeroom.

CENTER STUDIES

The Center has received nine special study assignments from AEC and one from ORNL. The assignments and their dispositions are as follows:

Contract File. – The Center is to prepare and maintain an active file of all nuclear safety contracts. The file will be used by the AEC as an active management tool and by the Center as a guide for its specialists. A preliminary draft of the file will be issued in July 1963.

Effect of Particle Agglomeration on the Penetrations of Filters Utilized with Double Containment Systems. – This study has been completed and a report issued.¹ In the study a simplified model was analyzed in which particles less than 0.3μ were assumed to penetrate the filters.

Environmental Hazard of Radioiodine from Reactors and Waste Effluents. – The Center will prepare a proposal on the effort needed to prepare a handbook on radioiodine which covers all pertinent phenomena from its generation to its ultimate exposure to man. The proposal will be submitted to the AEC in the fall of 1963. In the meantime, K. E. Cowser will conduct a more limited investigation on a related problem, namely, that of examining existing practice at Hanford, Idaho Falls, and Savannah River as regards iodine monitoring at the point of release and in the surrounding area.

Control and Safety System Appraisal. – An investigation that would, among other things, study the optimization of reliability vs redundancy has been assigned to C. S. Walker, and a report will be prepared within the next year.

Effect of Fuel Element Cladding Thickness in Water Reactors. – W. K. Ergen will assume responsibility for a study in this area to be issued in the fall of 1963.

Control Element Effectiveness Evaluations. – This problem has specific safety implications involving primarily reactor physics calculations and is largely outside the scope of the Center. A survey of the subject was conducted for an article² which appeared in *Nuclear Safety* last year. In view of the above, no further work is contemplated on this topic.

¹M. H. Fontana and W. E. Browning, Jr., *Effect of Particle Agglomeration on the Penetrations of Filters Utilized with Double Containment Systems*, ORNL-NSIC-1, Nuclear Safety Special Report, 1963.

²C. A. Preskitt, *Nuclear Safety* 3(3), 32-34 (1962).

Effectiveness of Safety Injection Systems for Emergency Reactivity Control. — This study has been assigned to C. S. Walker, and a study will be prepared within the next year.

Kinetics of Large Reactor Cores. — This study has been assigned to W. K. Ergen and P. H. Pitkanen, and a report will be prepared within the next year.

Review of Safety Analysis Procedures and Methods. — This subject is actually broader than

the six subject areas being initially covered by the Center. However, the general background and competence of persons associated with the Center will be used to fill uncovered areas. A report on the study will be issued within the next year.

Safety of Fast Gas-Cooled Reactors. — The ORNL Reactor Division has requested that the Center assess the status of the safety of fast gas-cooled reactors. The study has been assigned to W. K. Ergen, and a preliminary report is expected in the fall of 1963.

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Part V

Radiochemical Plants

Chemical Technology Division

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13. Radiochemical-Plant Safety Studies

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The hazards associated with a large radiochemical plant either for fuel processing or radioisotope source production may exceed those of a reactor. There are large amounts of fission products and/or nuclear materials in storage in such plants in readily dispersible form. Uncontrolled chemical reactions, fires, and nuclear accidents provide possible means for dispersal. It is imperative, therefore, that the hazards and means of combating them be assessed and understood before such a plant is built. In order to accomplish this, three general areas must be covered: (1) assessing the credible radiochemical-plant accidents and their consequences and determining which variables have a major effect on the hazards; (2) studying in detail the significant variables (e.g., the properties of aerosols produced in an accident, the mechanisms leading to a criticality accident, the maximum criticality accident possible, etc.); and (3) developing means for combating or ameliorating the consequences of radiochemical-plant accidents.

The analysis of radiochemical-plant hazards has been completed. An analysis of the hazards associated with spent-fuel processing plants was previously reported.¹ During this latest period a comprehensive survey of the hazards associated

with U^{233} and Pu^{239} fuel fabrication and radioisotope processing plants was made. This study served the dual purpose of (1) analyzing fuel fabrication and radioisotope processing plant accidents and (2) assessing the potential liability which might result from such accidents. The discussion presented here is essentially a summary of the study, which is reported in detail elsewhere.²

THEORETICAL POSSIBILITIES AND CONSEQUENCES OF MAJOR ACCIDENTS IN U^{233} AND Pu^{239} FUEL FABRICATION AND RADIOISOTOPE PROCESSING PLANTS

If the anticipated expansion in radioisotope sources and plutonium and U^{233} fuel for reactors is realized, several radiochemical plants for their production must be built and operated within the next 20 years. Typical 1980 yearly production is

¹C. E. Guthrie, E. D. Arnold, and J. P. Nichols, *Nuclear Safety Program Semiann. Progr. Rept. Dec. 31, 1962*, ORNL-3401, pp 63-72.

²C. E. Guthrie and J. P. Nichols, *Theoretical Possibilities and Consequences of Major Accidents in U^{233} and Pu^{239} Fuel Fabrication and Radioisotope Processing Plants*, ORNL-3441 (in press).

expected to be 10^4 kg of Pu^{239} fuel, 10^8 curies of Sr^{90} sources, and 100 kg of Pu^{238} isotope sources. Radiochemical plants are normally designed so that in the event of an accident one or more containment systems operate to limit the release of radioactive material to the environment (contained accident). A simultaneous failure of containment would result in the radioactive material being exposed to the environment (uncontained accident). The simultaneous occurrence of a fire or explosion that disperses the maximum amount of material, containment failure, and unfavorable meteorological conditions (worst uncontained accident) has such an extremely low probability of occurrence as to be deemed incredible. Even so, a study of these accidents is valid in order to obtain an upper limit on liability. The final results of this study are summarized in Table 13.1. This presents the potential economic loss from the worst contained and worst uncontained accidents in Pu^{239} , Sr^{90} , and Pu^{238} fabrication plants. The worst uncontained accident in such plants would possibly result in billions of dollars of economic loss due to damage to the surrounding land and population.

There has been no major off-site contamination from radiochemical-plant accidents to date. Although there is not sufficient radiochemical-plant operating experience to establish a probability for a major accident, in general, the frequency and severity of accidents in such plants has been lower than in the related chemical industry. A review of the accident experience to date does serve to point out that in most cases serious accidents result from an unforeseen combination of several independent circumstances and that relatively minor accidents can start a series of events that result in a major accident.

In order to evaluate the potential economic loss from radiochemical-plant accidents, contamination and personnel exposure levels were assumed and a dollar-loss value assigned to each level. These are summarized in Table 13.2. The contamination levels for I^{131} , Sr^{90} , and Cs^{137} were based on plant uptake studies. For the other isotopes, where such data are not available, the contamination levels were based on the MPC and a resuspension factor.

A population density increasing from zero at the site to 500 persons per square mile at 20 miles and farther from the site was found to be reasonably typical for fuel fabrication and radioisotope processing plants. This is essentially the same distribution used in the comparable reactor liability study.³ A uniform population density of 100 persons per square mile was also used in calculating the loss economics to allow facile conversion of the results to various sites.

The area and downwind distance as a function of isopleths of exposure and ground contamination were calculated with the Gaussian plume-dispersion model, using experimentally verified values of dispersion coefficient, velocities conducive to deposition of particles (deposition velocities), and washout rates for practical, consistent sets of weather conditions.⁴ The effect of weather conditions on the loss during uncontained accidents is summarized in Table 13.3. Washout maximizes contamination areas, and inversion maximizes

³Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants, WASH-740, p 78 (March 1957).

⁴W. M. Culkowski and W. F. Hilsmeier, Maximum Values of Concentration, Fallout and Washout for Radioisotope Releases, ORO-599 (March 1963).

Table 13.1. Summary of Potential Economic Loss in Worst Contained and Worst Uncontained Accidents

Type of Plant	Worst Contained Accident		Worst Uncontained Accident	
	Release to Atmosphere	Potential Economic Loss	Release to Atmosphere	Potential Economic Loss
Pu^{239} fuel fabrication	40 g (2.5 curies)	$\$3 \times 10^6$	4 kg (250 curies)	$\$4 \times 10^8$
Sr^{90} source fabrication	220 curies (1.5 g)	2×10^7	50,000 curies (350 g)	5×10^9
Pu^{238} source fabrication	2.2 g (37 curies)	5×10^7	2 kg (34,000 curies)	5×10^{10}

Table 13.2. Summary of Monetary Loss Assumptions and Limits Assumed for Personnel Exposure and Area Contamination

Loss Category	Effect	Assumed Loss Per Person	Sr ⁹⁰	Pu ²³⁹	Pu ²³⁸
Personnel Exposure to Critical Organ (rems per 13 weeks)			Exposure Limit (curies sec m⁻³)		
A	>500	\$50,000	>4.1	>6.3 × 10 ⁻²	>6.3 × 10 ⁻²
B	>50 but <500	10,000	>4.1 × 10 ⁻¹ to 4.1	>6.3 × 10 ⁻³ to 6.3 × 10 ⁻²	>6.3 × 10 ⁻³ to 6.3 × 10 ⁻²
C	>5 but <50	2,000	>4.1 × 10 ⁻² to 4.1 × 10 ⁻¹	>6.3 × 10 ⁻⁴ to 6.3 × 10 ⁻³	>6.3 × 10 ⁻⁴ to 6.3 × 10 ⁻³
Contamination			Contamination Limit (curies/m²)		
I	Severe (long-term evacuation; total loss of value; no crops ≥5 years)	\$10,000	>1.1 × 10 ⁻⁵	>6 × 10 ⁻⁷	>7 × 10 ⁻⁷
II	Moderate (short-term evacuation; extensive decontamination; no crops 1-5 years)	1,500	>1 × 10 ⁻⁶ to 1.1 × 10 ⁻⁵	>6 × 10 ⁻⁸ to 6 × 10 ⁻⁷	>7 × 10 ⁻⁸ to 7 × 10 ⁻⁷
III	Minor (no evacuation; minor decontamination; some crops destroyed)	0.005 ^a	>1 × 10 ⁻⁷ to 1 × 10 ⁻⁶	>6 × 10 ⁻⁹ to 6 × 10 ⁻⁸	>7 × 10 ⁻⁹ to 7 × 10 ⁻⁸

^aPer m² (not per person).

Table 13.3. Effect of Weather on Economic Loss Due to an Accident Involving a Release of 50,000 Curies Sr^{90}

Loss Category	Potential Economic Loss		
	Inversion	Washout	Sunny Day
I	1.0×10^9	1.6×10^9	9.0×10^5
II	2.3×10^9	3.0×10^9	5.8×10^5
III	4.0×10^8	4.2×10^8	6.0×10^4
A	5.0×10^7		
B	1.0×10^7	1.6×10^4	2.8×10^4
C	1.3×10^6	1.6×10^5	5.2×10^4
Total	3.8×10^9	5.0×10^9	1.6×10^6

personnel exposure. In most cases the maximum loss is for the washout condition.

The consequences of accidents involving 17 isotopes have been calculated and the potential economic loss presented as a function of curies released for each. The potential loss as a function of Sr^{90} release is shown in Figs. 13.1 and 13.2. The quantity of material that must be released to cause a loss of \$60 million is presented in Table 13.4 for several isotopes. Release of the fission products from a criticality accident or the worst uncontained accidents involving U^{233} or Kr^{85} would result in less than \$60 million in damage.

The worst uncontained accident in a Pu^{239} fuel fabrication facility was postulated to result from a

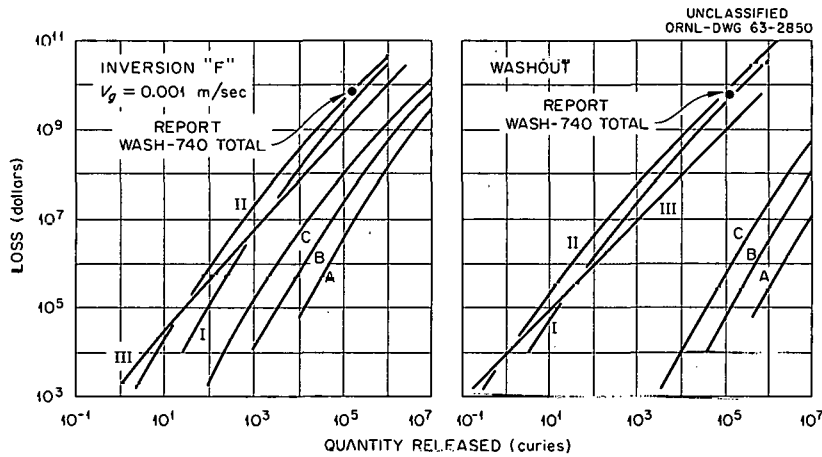


Fig. 13.1. Potential Economic Loss Resulting from Release of Sr^{90} . Typical population distribution.

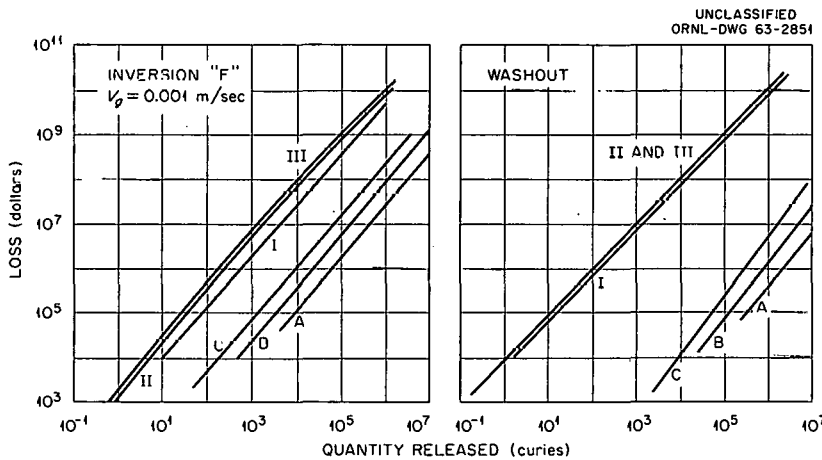


Fig. 13.2. Potential Economic Loss Resulting from Release of Sr^{90} . One hundred persons per square mile.

fire and carbon monoxide explosion in a storage area for graphite fuel elements and resulted in the release of 4 kg of Pu²³⁹ (4% of the inventory) as smoke to the atmosphere. The worst uncontained accident in an isotope recovery and source fabrication plant was assumed to result from a solvent explosion in the cell, dispersing 20% of the activity to the atmosphere. These and other postulated accidents are more fully described and the potential economic losses (Table 13.1) are presented in detail elsewhere.²

Table 13.4. Quantity of Material Released to Cause \$60 Million in Damage Under Washout Conditions

Isotope	Amount Released	
	(g)	(curies)
Pu ²³⁹	1000	60
Pu ²³⁸	3	60
Sr ⁹⁰	7	1,000
I ¹³¹	0.6	80,000

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Part VI Space Safety

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14. Environment Hazards

RADIOLOGICAL SIGNIFICANCE OF NUCLEAR-ROCKET DEBRIS

T. G. Clark B. R. Fish
S. A. Lewis¹ P. A. Thompson¹

A preliminary review has been made of the potential biological hazards from deposition of nuclear-rocket debris in the earth's biosphere.² In that review the suborbit mode of nuclear-rocket operation is considered with respect to the potential hazards associated with reactor fragments produced by explosive destruction of the nuclear rocket. Reactor debris particles ranging in size from 1 cm down to respirable sizes have been considered with respect to the biological effects resulting from inhalation, ingestion, external radiation from ground deposits, and beta irradiation of the skin in contact with such particles. Although the available data are not entirely adequate for the assessment of potential hazard, the preliminary review suggests that the most significant radiological aspect is the intense beta irradiation from relatively insoluble particles surviving reentry. In view of the potentially high levels of radiation exposure indicated by this preliminary review, a more refined analysis has been undertaken for other anticipated types of operating and potential accident situations. The major question at this time

¹Temporary summer employee, Yale University, New Haven, Conn.

²B. R. Fish and G. R. Patterson, Jr., ORNL TM-501 (classified).

is the degree and the significance of intense beta radiation emitted by reactor debris particles.

In a recent report by E. V. Hulse³ of the Radio-biological Research Unit at Harwell on the production of tumors in mice following external beta irradiation, mention is made of extrapolating the results to man. This follows the principle that in the absence of sufficient human data the safest course in a hazards evaluation would be to use information from the most sensitive experimental animal.

In this country some of the most pertinent current work on the "hot-particle" problem is being conducted by Dr. Roy E. Albert, New York University College of Medicine. Discussions with this investigator affirmed the consensus that the reaction of skin (whether of man or animal) to beta rays is most likely a function of some as yet undetermined variation in relative sensitivity of different histological units. Thus, the distribution of hair follicles, sebaceous glands, etc. per unit of skin may be of controlling importance.

Good animal data will be checked against available human data to see if this type of extrapolation is valid. A literature search is under way for the purpose of comparing human beta-ray exposures with available animal data. Relatively few cases of external beta irradiation which include adequate dosimetry have been documented, and, for the most part, these stress the acute effects. An independent survey of possible latent effects has been

³E. V. Hulse, *Brit. J. Cancer* 16(1), 72-86 (1962).

initiated. The subject group consists of several people who voluntarily exposed limited areas of skin to beta radiation from P^{32} plaques. The radiation was given approximately 18 yr ago in graded doses, the maximum being 1180 rads. Another source of information regarding delayed response of human skin to beta irradiation is R. A. Conard, Brookhaven National Laboratory, who has been consulted regarding his studies of the Marshallese. Of special interest was the appearance six years post exposure of benign pigmented macules in the area of healed beta burns.

A computer program is being written to calculate the dose delivered to tissue from a small fragment

of the reactor core following a power cycle, destruction, and fallout. The primary beta and gamma radiation from the fission products are divided into a finite number of energy bands which are treated as vectors. The degradation of each vector as it proceeds through matter will be determined by an iterative process. At present the major effort is being directed toward computing the cross sections which make up a "degradation matrix." A cross-section program has been written and has produced generally satisfactory results although some portions of these programs are still being debugged. Only preliminary work has been done on the main program.

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