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SEMIANNUAL PROGRESS REPORT
FOR PERIOD ENDING DECEMBER 31, 1962

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 December 31, 1962

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SUMMARY

1. Introduction

This is the second in a series of semiannual reports describing the Nuclear Safety Research and Development Program at the Oak Ridge National Laboratory. The first report in this series, ORNL-3319, was published in August 1962.

PART I. PHYSICS

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

It was concluded from preliminary results that two-dimensional diffusion-theory codes for calculating reactivity effects of small fuel displacements in pool-type reactors were not adequate for the types of small-geometry, large-cross-section perturbations produced in the first experiments.

A slab-reactor experiment is planned to test the adequacy of transport calculation methods. It will be performed at the Critical Experiments Facility at ORNL, with fuel elements that have been obtained from the AEC.

The later phase of the experimental program has been deferred until enough proper fuel plates are available.

PART II. CHEMISTRY

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

Fission-product-release data have been obtained by use of two new techniques which permit melting of larger amounts of irradiated UO_2 than

was possible with previously available techniques. The two scaled-up procedures gave results which are essentially in agreement and which confirm most of the data obtained earlier with milligram amounts of UO_2 irradiated at tracer level. The first new method, melting of UO_2 contained in a tungsten crucible surrounded by a helium atmosphere by using rf heating, gave essentially quantitative release of rare gases and iodine but somewhat lower releases of tellurium and cesium. Essentially no ruthenium or strontium was released. The most apparent explanation of the lower release values observed, as compared with those obtained in earlier small-scale experiments, is that the greatly increased volume-to-surface ratio prevailing in these experiments resulted in a longer diffusion path for the fission products. The lack of oxygen in the helium atmosphere also helped to reduce ruthenium volatility.

The second method, melting of cored UO_2 pellets surrounding a tungsten resistor, permitted a comparison of the effect of two different metal cladding materials on fission product release. It was found that stainless steel cladding simply segregated and had little effect on release as compared with that observed from unclad UO_2 . Molten Zircaloy, however, apparently wet the UO_2 , producing a decreased tellurium release by a factor of 100 and an increased strontium release by a factor of almost 10. A possible interpretation of this phenomenon is that the molten zirconium spreads over the surface of the UO_2 and serves as a getter for excess oxygen and for metals such as tellurium, with which it alloys readily. The lower availability of oxygen would be expected to result in increased volatility of strontium, because the metal is more volatile than the oxide. It was also noted that the usual ratio of 2 to 3 times as much barium as strontium release was reversed in this case.

Hot-cell construction is under way to permit the melting of larger quantities of irradiated UO_2 in tungsten crucibles and the clad UO_2 specimens currently being irradiated. A new ultra-high-frequency generator has been tested for use in both programs.

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

The series of experiments in the ORR previously described is being continued to study the amounts and forms of fission products released

during simulated reactor accidents. The first type of experiment consists of melting or vaporizing a miniature stainless-steel-clad UO_2 fuel element in a helium atmosphere. The second type consists of burning in air a miniature fuel element composed of spheroidal particles of uranium carbide coated with pyrolytic carbon and embedded in a graphite matrix. In each case, fission and gamma heat raised the temperature of the fuel element sufficiently to cause destruction.

In the experiments with UO_2 , nearly all the I, Te, and Cs and more than half the Sr, Zr, Ru, Ba, and Ce were released from the fuel. Release of the latter group of fission products and uranium from the high-temperature zone is generally less than 3%. This decrease in release values from fuel to high-temperature zone is considered to be significant since during an actual reactor accident temperatures corresponding to those of the high-temperature zone would probably occur only within the reactor containment.

In the experiment with UC_2 -graphite, fuel release values were lower except for ruthenium, which probably formed volatile ruthenium oxide.

One objective of this program is to determine the characteristics of the fission products which are released. Analysis of the observed distribution of the various fission products along the wall of the exit-gas tube shows that they are apparently associated with two particle-size groups in the millimicron range in the UO_2 experiments. 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 knowledge of fractionation mechanisms will aid in the recognition of the relative importance of the various factors which define a reactor accident.

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

In preparation for a series of experiments on the release of fission products from UO_2 melted under reactor transient conditions, to be conducted in the TREAT facility at the NRTS, an initial program proposal was reviewed with TREAT management for the purpose of reaching an agreement on the design of the experimental assembly.

The proposed experiments will permit the study of fission product release on melting fully clad, 10%-enriched UO_2 in an argon-filled autoclave by means of fast transients of varying magnitude. Provision will be made for preheating the samples to 1200°C and for sampling the released gases and smoke promptly by admission through a filter into an evacuated autoclave. The fuel specimens in the first experiments in this facility will not be irradiated prior to melting, but specimens having a high degree of burnup will be melted later. Heat transfer determinations and leakage testing of the autoclaves have been satisfactorily completed. A hazards analysis is presently being prepared, and the first experiment is expected to be inserted in the reactor during the early part of 1963.

6. Fission Product Transport Evaluations

Two methods for determining the heat of vaporization of radioactive species have been tested with I^{131} -labeled iodine in the search for a technique to identify the chemical form of fission products released in simulated reactor accident experiments. One method, which depended on the rate of transfer of iodine by helium flowing at a constant rate, gave values for the heat of vaporization of iodine ranging from 13 to 15 kcal/mole. The average value, 14.2 kcal, agreed reasonably well with the literature value of 14.9 for solid iodine. A completely sealed glass system, consisting of two small bulbs connected by a long capillary tube comprised the sample container employed in the second method. The range in concentration of iodine vapor in one bulb, which was in equilibrium with solid or liquid iodine in the other bulb, was measured over a range of temperatures by a crystal detector. Early results obtained by this method varied over a wider range than those obtained by the transport method; however, recent data indicate that this may be a useful technique, and experiments are in progress to obtain a better understanding of iodine behavior in this apparatus. Evidences of adsorption of iodine on glass or quartz walls were observed in both types of experiments.

7. Characterization and Control of Accident-Released Fission Products

Two methods for determining the form of radioactivity in gases have been investigated. In the first method for characterizing aerosols, diffusion coefficients for fine particles and for radioactive vapors such as iodine are determined by measuring the distribution of radioactivity on the walls of a channel previously exposed to gas carrying radioactive materials and flowing under laminar conditions. This method has been applied to the problem of removing millimicron-diameter radioactive particles from air streams. In tests using I^{131} -labeled Al_2O_3 particles with a nominal diameter of 0.004μ , high-efficiency, low-pressure-drop particulate filters exhibited an efficiency ranging from less than 10% up to about 75%. With activated-carbon beds, 0.75 in. deep and 6×16 mesh, iodine vapor removal was essentially complete, but the removal efficiency for 0.003- to $0.006\text{-}\mu$ particles was observed to be only 75-90%, the higher efficiency being associated with the finer particles. Similar tests were carried out using fog condensation and foam encapsulation to remove millimicron-sized particles from air. In these tests removal efficiencies of 85 to 99% were observed.

The second method for measurement of the characteristics of radioactive aerosols makes use of fibrous filters. This method measures aerosols in terms of their response to the processes of inertial impaction, interception, and diffusion under carefully controlled conditions. A filter was developed having uniform fiber diameter to permit theoretical analysis and having a layered structure to facilitate separation of the fiber bed into discrete layers for radioassay after exposure to the aerosol. Preliminary results with a radioactive aerosol of 0.004- to $0.03\text{-}\mu$ particles, containing Zn^{65} , indicate that the techniques used are satisfactory and should provide useful information on aerosol character and on filtration mechanisms.

PART III. CONTAINMENT ENGINEERING

8. 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 installation is virtually complete, with delivery of most of the components scheduled for early in 1963. The outstanding design work is concerned with remote-handling tools and casks for transporting components and samples. Arrangements were made to conduct an experiment in the NSPP cell (prior to the installation of NSPP components) to measure the effectiveness of foam in trapping released activity. This experiment is now in progress using 10 curies of I^{131} dispersed in the atmosphere of the 6000-ft³ cell.

X 9. Reactor Containment Handbook

A handbook for use in reactor containment system design is being prepared by ORNL and subcontractor, Bechtel Corporation. An outline of the handbook is given, together with the estimated percent completion of the rough draft of each chapter. In all, draft material for approximately 20% of the handbook is prepared with expected completion scheduled for 1963.

PART IV. RADIOCHEMICAL PLANTS

10. Radiochemical-Plant Safety Studies

An evaluation was made of the hazards associated with the maximum credible accidents in a 1-ton/day spent-fuel processing plant. The major conclusions are as follows: (1) As long as the absolute-filter systems retain their 99.97% efficiency, there is negligible hazard if both primary and secondary containment hold, only minor hazard if one of the two containments fails, and a major hazard (contamination of $> 10^5$ m²) if both containments fail. (2) The hazards associated with an absolute-filter

failure are dependent on meteorological conditions. Under the worst conditions (washout) a filter failure could be just as serious (contamination of up to 10^6 m²) as a failure of both containment systems. Experience at ORNL with degradation of absolute-filter systems in chemical-plant service, in connection with these results, accentuates the vital need to develop a chemically resistant absolute filter.

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PART IV. RADIOCHEMICAL PLANTS

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1. INTRODUCTION

This is the second 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 four 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; and, fourth, collecting, critically evaluating, and compiling, in handbook form, information on reactor containment. 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 fraction of the fission-product inventories of important reactor fuels which may be released 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 containment 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.

The first report in this series, ORNL-3319, was published in August 1962.

PART I PHYSICS

NEUTRON PHYSICS DIVISION

E. P. Blizard, Division Director
F. C. Maienschein, Coordinator

2. REACTIVITY EFFECTS OF SMALL FUEL DISPLACEMENTS IN A POOL-TYPE REACTOR

E. G. Silver

As was reported in the previous summary,^{1,2} a program to determine the reactivity effects of small fuel displacements in pool-type reactors is under way.

Preliminary experimental and calculational results were reported, which led to the conclusion that two-dimensional diffusion-theory codes could not adequately handle the types of small-geometry, large-cross-section perturbations produced in the first experiments.

In order to test the adequacy of transport calculation methods for calculating this type of perturbation, using the available one-dimension transport code (DSN), a slab-reactor experiment is planned to compare with such calculations.

The fuel elements for this test have been obtained on loan from the AEC and have been shipped to the Critical Experiments Facility at ORNL, where the experiments will be performed.

The fuel plates are 58 mils thick, 2.95 in. wide, and 72 in. long and are made of unclad uranium-aluminum alloy material employing highly enriched U^{235} . Each plate contains 58.6 g of U^{235} , in a 10.5 wt % alloy. Ten of these plates will be combined into a "fuel element" 58 mils thick,

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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.

29.5 in. wide, and 72 in. long. These plates will be stacked, standing with appropriate water spacing in a parallelepipedal tank in which water height will serve as reactivity control.

The tank is so arranged that, when the water height is just equal to the vertical dimension of a fuel element, the reactor is reflected only on the two faces adjacent to the end fuel-element surfaces and is bare on the four faces adjacent to the fuel-element edges. The thickness of the stack will be adjusted by adding fuel elements at the ends, with partial elements if needed, to achieve criticality with this exact water height. The perturbations are then performed by selectively removing or adding elements near the core center, and compensating by either adding or subtracting fuel, or by making small, calibrated water-height changes.

The later phase of the experimental program, as outlined in the previous report, utilizing a "three-dimensional" core of fuel plates of Bulk Shielding Facility dimensions (2.9 in. by 60 mils by 24 in.) in a core with flexible plate spacings, has been deferred because a sufficient number of loose fuel plates with precise enough fuel-weight and fuel-dimension tolerances are not available.

PART II CHEMISTRY

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

Studies of fission product release from irradiated UO_2 are being made under out-of-pile conditions to permit maximum freedom in designing and conducting experiments. Emphasis is currently being placed on development of methods and equipment to permit melting of large quantities of irradiated UO_2 under conditions more realistic than those employed in earlier UO_2 melting experiments. Preliminary results on the effect of cladding and length of time in the molten condition on the release of fission products from melted UO_2 were obtained.

FISSION PRODUCT RELEASE FROM MELTED UO_2 BY THE TUNGSTEN-CRUCIBLE METHOD

A brief description of the tungsten-crucible method for melting UO_2 was given in the previous progress report.¹ One of the principal advantages of this method is that it can be scaled up to any desired batch size, but it was convenient to melt about 30 g of UO_2 under the prevailing experimental conditions. Other advantages include the ability of tungsten to contain molten UO_2 , which permits an extended molten period without

¹G. W. Parker et al., Nuclear Safety Progress Semiann. Rept., June 30, 1962, ORNL-3319, pp 11-29.

loss of material, and the adaptability of the tungsten-crucible method to the use of thermocouples for temperature measurements. The fact that it is limited to an inert-gas atmosphere is not regarded as a serious disadvantage, since it has already been shown that the effect of atmosphere on release is minor at the melting temperature of UO_2 . Studies of the effect of atmosphere are being continued by the use of two other methods for melting fuel.

The program has been extended to include experiments in which trace-irradiated UO_2 pellets were melted and held molten for a longer period than in previous work as part of a series of experiments to investigate the effect of duration of the molten period on fission product release. The rf furnace and the fission-product-fractionation assembly are shown in Fig. 3.1. A thin-walled tungsten crucible inside a small thorium oxide crucible, which served as a heat reflector, was used as the heater and container. Helium gas, purified by passing it over hot zirconium, flowed upward through the furnace at a rate of $700 \text{ cm}^3/\text{min}$, measured at room temperature. A typical melt is shown in Fig. 3.2. The released solids were divided into three major fractions for analysis: (1) those plated out in the furnace tube, (2) those deposited on the wall of a miniature expansion chamber, and (3) gas-borne solids collected on a Millipore filter. The solids in the different parts of the apparatus were dissolved, and the solutions were analyzed. Release values were calculated from a complete material balance. Charcoal traps were also employed to collect molecular iodine and the rare gases. For chemical recovery of the fission products remaining in the melt, the tungsten crucible was placed in a nitric acid solution, which dissolved the UO_2 .

Release as a Function of Time in the Molten State

One question which has provoked much speculation concerns the effect of the length of time a sample is kept above the melting temperature. Little freedom in extending this period was possible before the tungsten-crucible method was developed. Consequently, this is the first variable to be investigated. Data obtained in the first experiments are given in Table 3.1. Relatively little can be said about these results, which are

Fig. 3.1. Furnace and Fission-Product-Collection Equipment for Melting UO_2 by the Tungsten-Crucible Method.

Fig. 3.2. Tungsten Crucibles and UO_2 After Melting.

quite tentative, but it appears that scaleup has served to reduce some of the release rates, especially those of tellurium and cesium. Some of the data in Table 3.1 seem to indicate an increased release with increased molten time, but insufficient data are available to be conclusive on this point. The materials deposited in different parts of the apparatus were analyzed separately, and the results are summarized in Table 3.2. It will be of interest to conduct melting experiments with similar amounts of highly irradiated UO_2 when suitable hot-cell facilities become available.

Size of Uranium Oxide Particles Plated Out from Molten UO_2

A size analysis was performed on the material which appears to be released as a vapor rather promptly on melting UO_2 . In practice it is useful to watch for the oxide smoke to rise above the sample as an indication

Table 3.1. Fission Product Release from UO_2 Melted in Helium
by the Tungsten-Crucible Method

Sample size: 29 g of PWR UO_2 , trace-irradiated
Atmosphere: Purified helium, $700 \text{ cm}^3/\text{min}$

Date of Run	Time (min)		UO_2 Vaporized (%)	Release (%)								
	Preheat	Molten		Gross Gamma	Rare Gas	I	Te	Cs	Ru	Sr	Ba	Ce
10-24-62	4.75	1.0	0.097	22.6	92.9	98.1	81.0	62.5	0.45	0.33	4.8	0.05
10-31-62	5.0	1.5	0.157	15.1 ^a	98.0	98.3	82.8	65.0	0.05	0.47	2.57	0.07
11-7-62	4.75	2.0	0.156	25.6	99.3	98.8	99.1	59.5	0.32	0.41	2.95	0.17
11-12-62	4.5	2.5	0.246	12.6 ^a	99.0	95.0	98.9	72.0	0.33	0.53	2.4	0.13
11-15-62	4.5	1.5		13.7 ^a	98.7	88.4	91.8	80.2	0.20	0.26	2.6	0.40
11-19-62	4.5	2.5		12.6 ^a	99.3	92.9	96.2	89.4	0.70	0.50	3.6	0.10

^aDecayed one week longer.

Table 3.2. Distribution of Released Fission Products from UO_2 Melted
in Helium by the Tungsten-Crucible Method

Location	Release (%)									
	Total Gamma		Iodine		Tellurium		Cesium		Strontium	
	10/24	10/31 ^a	10/24	10/31	10/24	10/31	10/24	10/31	10/24	10/31
Furnace	17.8	11.6	45.3	64.1	62.9	68.5	51.6	43.6	0.2	0.4
Fallout chamber	0.8	1.1	2.8	13.5	4.7	3.0	4.5	15.8	0.1	0.04
Filter	2.8	2.3	8.0	18.1	13.4	11.3	6.4	6.8	0.02	0.04
Charcoal	1.2	0.1	42	2.8						
Total	22.6	15.1	98.1	98.5	81.0	82.8	62.5	66.2	0.32	0.48

^aDecayed one week longer.

that melting has actually occurred. Much of the vaporized oxide plated out in the furnace tube just above the crucible. A fraction of the vapor was carried into the expansion chamber, where most of it deposited on the bottom. A filter was inserted beyond this chamber to stop all remaining solids. Samples from these areas were examined and photographed. The progression from the furnace tube (Fig. 3.3) to the front (Fig. 3.4) and back (Fig. 3.5) of the expansion chamber and then to the filter (Fig. 3.6) shows (1) a mixture of sizes in the furnace tube, (2) a slightly larger particle size in the fore part of the chamber than in the rear, and (3) a still smaller size on the filter. A median 50% size varied only from 0.03 to 1.0 μ for all samples. A count analysis is given in Fig. 3.7.

MELTING OF UO_2 FUEL PINS BY A CENTERED TUNGSTEN RESISTOR

Nuclear melting has been more directly simulated by use of center heating with a tungsten resistor (Fig. 3.8) surrounded by cored UO_2 pellets than was possible with previously available techniques. Experiments in which unirradiated UO_2 was melted in a helium atmosphere were reported previously.¹ Fission product release has recently been investigated by use of the new method, in which the amounts of UO_2 melted were about 1000 times the quantity melted in the arc-image furnace. The experimental assembly employed in these experiments is shown in Fig. 3.9. It is of interest to compare results obtained by the use of two methods and to study the effect of another variable, metal cladding.

Comparison with Previous Melting Experiments

In Table 3.3, the results obtained on melting three unclad specimens and three metal-clad specimens, one of stainless steel and two of zirconium alloy, are summarized. In comparing these results with those from the microsample (~30-mg) experiments, it is apparent that two differences in experimental conditions must be taken into consideration: (1) the oxygen present in the impure helium employed in the former work apparently caused a high ruthenium release by oxidizing Ru to RuO_4 ; (2) incomplete melting of the center-heated specimens resulted in incomplete rare-gas release. It is, in fact, probable that the percent of gas release is actually a measure of the percent of the sample melted.

Fig. 3.3. UO_2 Particles Collected on Furnace-Tube Wall. 26,000X.

Fig. 3.4. UO_2 Particles Collected in Fore Part of Fallout Chamber. 26,000X.

Fig. 3.5. UO_2 Particles Collected in Rear Part of Fallout Chamber. 26,000X.

Fig. 3.6. UO_2 Particles Collected on the Backup Millipore Filter Paper. 26,000X.

Fig. 3.7. Particle-Size Evaluation of Oxides from UO_2 Samples Melted in Helium by the Tungsten-Crucible Method.

Fig. 3.8. Single-Element Tungsten-Resistor Furnace.

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Fig. 3.9. Furnace Assembly and Fission-Product-Release Train for Tungsten-Resistor Melting Method.

Table 3.3. Fission Product Release from Single-Element UO_2 Melted
in a Tungsten-Resistor Furnace

Sample: 39 g of PWR UO_2 , trace-irradiated
Atmosphere: Pure helium flowing at a rate of $400 \text{ cm}^3/\text{min}$

Element	Heat Duration (min)	UO_2 Vaporized (%)	Release (%)								
			Gross Gamma	Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Ce/RE
UO_2	5.0	0.8	7.1	63	47	56	44	1.6	1.6	5.3	< 0.6
UO_2	4.0	0.2	5.7	50	30	42	41	0.4	0.9	2.9	< 0.5
UO_2	4.4	0.3	6.9	41	25	29	> 40	0.03	1.2	4.3	0.5
UO_2 (SS clad)	4.7	0.2	5.0	56	52	73	46	0.5	1.0	3.4	0.1
UO_2 (Zr clad)	7.0	0.1	2.6	52	24	1.9	27	0.08	11.6	7.7	0.5
UO_2 (Zr clad)	6.7	0.04	5.2	41	50	0.6	32	0.2	9.9	7.5	0.5
$\text{UO}_2 + \text{BeO}^a$	1.5	20	30	100	90	92	91	61	2.1	4.6	2.2

^aPrevious results of melting microsize sample of highly irradiated UO_2 in impure helium by use of the carbon-arc image method.

Effect of Cladding Material on Fission Product Release

The presence of cladding material during melting experiments was not expected to have any effect on fission product release. Melting UO_2 in contact with stainless steel resulted in a heterogeneous segregated residue (Fig. 3.10), whereas Zircaloy apparently wet the molten UO_2 and formed a solid solution (Fig. 3.11). It is not especially surprising that some effect of this wetting action of Zircaloy was noted in the release data. A remarkable reduction of the release of tellurium by a factor of almost 100 was observed. A slight reduction in cesium and iodine release also seems to be indicated by the data obtained. This advantage is offset by a serious increase of strontium release from 1% to 10%. Barium release also increased from 3% to 7%. This effect is probably related to oxygen depletion of the system by hot zirconium, but it requires further attention. Very little UO_2 appeared to have vaporized and deposited on nearby surfaces, and only trace amounts reached the filter. With these exceptions, and that of the behavior of ruthenium attributed to impure helium, the microsize samples appear to have given valid release data, as compared with values obtained with 39-g samples. Enough uranium oxide reached the filter in some experiments to permit a particle-size analysis. The sizes and distribution observed are consistent with those observed in similar experiments. Photographs of particles (Figs. 3.12 and 3.13) and a particle distribution analysis (Fig. 3.14) illustrate the range of particle sizes encountered in these experiments.

A distribution analysis which serves to show the location of various deposited fission products is given in Table 3.4.

Scaleup of the Tungsten-Resistor Melting Method to a Multiple-Pin Assembly

One of the most important simulation concepts yet to be demonstrated on a scale intermediate between that of present laboratory experiments and full reactor size is the plate-out effect of fission products from a completely melted fuel pin surrounded by partially melted pins. The tungsten-resistor melting method offers a practical approach to this demonstration, and it has been scaled up in design to the seven-cluster assembly shown in Fig. 3.15. It is conceived as being fitted with six unirradiated pins on the outer circle and with only the center pin having

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Fig. 3.10. Appearance of Stainless-Steel-Clad Pin After Melting.

Fig. 3.11. Appearance of Zirconium-Clad Pin After Melting.

Fig. 3.12. Particles of Uranium Oxide Collected from Melting a Stainless-Steel-Clad UO_2 Fuel Pin. 8000X.

Fig. 3.13. Particles Collected from Melting a Zirconium-Clad UO_2 Fuel Pin. 8000X.

Fig. 3.14. Particle-Size Analysis of UO_2 Vaporized from Stainless-Steel-Clad and Bare UO_2 Fuel Pins.

Fig. 3.15. Tungsten-Resistor Furnace with Clustered Elements.

Table 3.4. Distribution of Released Fission Products from UO₂ Melted in Helium by Center Resistor Method

Location	Release (%)																	
	UO ₂			Iodine			Tellurium			Cesium			Strontium			Barium		
	Bare	SS	Zr	Bare	SS	Zr	Bare	SS	Zr	Bare	SS	Zr	Bare	SS	Zr	Bare	SS	Zr
Furnace hot zone	0.42	0.02	0.1	0.31	0.7	0.3	2	14	0.1	24	7	9	1.2	1.0	10.6	4.1	3.2	7.7
Base and gas tube	0.002	0.004	0.002	13.8	28	27	23	53	1.0	10	25	11	0.004	0.01	0.01	0.03	0.02	0.05
Filters	0.0001	0.004	0	11.2	23	9	8.7	5	0.1	12.3	14	5	0.001	0.006	< 0.01	0.002	0.002	0.006
Charcoal				1.0	0.2	0.4												

a fission product inventory. Examination of the outer pins after melting, then, will show where the fission products deposit after release from the adjacent hot fuel. A 25-kw saturable reactor has been obtained for use as the power supply, and a demonstration run will be conducted in the near future.

HOT-CELL CONSTRUCTION FOR MELTING MTR-IRRADIATED CAPSULES

Approximately 60 stainless-steel- and zirconium-clad capsules containing up to 80 g of UO_2 are being irradiated in the MTR to provide samples for an extensive hot-cell, enlarged-scale melting program. Irradiation of the capsules began August 6, 1962. Design work has been completed for most of the complex in-cell melting, handling, and chemical

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HORIZONTAL SECTION AT EL +12 ft-0 in.

Fig. 3.16. Hot-Cell Plan for Remote Handling and Melting of Clad UO_2 Fuel Capsules.

processing equipment. Much of the equipment has been constructed; however, some items are still in the development stage. Three separate cells will be required to accomplish the multiple operations necessary for the program. A schematic plan of equipment layout is given in Fig. 3.16.

A new 5-Mc induction generator will be used for melting irradiated fuel specimens in a hot cell. This permits heating by direct coupling with the oxide fuel itself, following decladding by induction coupling to the stainless steel or zirconium cladding material.

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4. RELEASE OF FISSION PRODUCTS ON THE IN-PILE MELTING OR BURNING OF REACTOR FUELS

W. E. Browning, Jr.
C. E. Miller, Jr.

B. F. Roberts
R. P. Shields

The location of nuclear power reactors near populated areas is desirable from an economic viewpoint but generally undesirable from the hazards viewpoint. The nearness of the site to a populated area depends on the extent to which these opposing views can be reconciled. One of the more important hazards is the release to the environment of fission products resulting from a nuclear accident. If complete release of all fission products is postulated, large exclusion areas must be provided around reactor sites. In-pile experiments are being performed at ORNL to study the release of fission products during simulated reactor accidents in order to provide reliable data as to the amounts and forms of fission products released, and thus to reduce the need for these large exclusion areas.

Two types of experiments have been conducted in the ORR to simulate reactor accidents in which fuel elements are destroyed by melting or burning. The first type consists of melting or vaporizing a miniature stainless-steel-clad UO_2 fuel element in a helium atmosphere. The second type consists of burning in air a miniature fuel element composed of spheroidal particles of uranium carbide coated with pyrolytic carbon and embedded

in a graphite matrix. In each case fission and gamma heat raised the temperature of the fuel element sufficiently to cause destruction.

The apparatus employed in these experiments and the results of visual examination of the first five UO_2 melting experiments were reported previously.¹ Eight melting experiments and one burning experiment have now been completed.

Experiments 6, 7, and 8 were a part of the series designed to determine the reproducibility of the UO_2 melting experiments. Each contained the same fuel as experiments 3, 4, and 5 (6.5% enriched UO_2), and conditions such as insertion rate and duration of melting were constant. Examination of these experiments in the hot cells showed that the specimen in experiment 6 was 88% melted, the specimen in experiment 7 did not melt but formed a central void similar to that produced in experiment 1, and the specimen in experiment 8 was 79% melted. In these three experiments variations in the reactor core configuration resulted in a low neutron flux and thus in low heat generation. The fuel residues in both experiments 6 and 8 were similar to those in experiments 4 and 5 — solid masses with apparent spattering of fuel on the side of the ThO_2 cylinder. Figure 4.1 shows the appearance of the fuel and fuel holder from experiment 6. The fuel residue from experiment 7 (Fig. 4.2) was crystalline and exhibited the same pyramid-shaped crystal structure described¹ for experiment 1.

Radiochemical analyses for various fission products have been performed for all eight melting experiments. With the exception of experiment 1, which was performed primarily to test equipment, the results are shown in Tables 4.1 and 4.2. Table 4.1 gives the percentages of various fission products released from the fuel. Each release value is based on the total amount of that fission product found throughout the experimental assembly. The data in Table 4.1, except for experiment 7, show, in general, that nearly all of the iodine, tellurium, and cesium were released, as well as more than half of each of the other fission products. One-third to one-half of the UO_2 was transferred from the holder to other parts of the high-temperature zone, and the fission products contained in

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

Fig. 4.1. Fuel and Fuel Holder from UO₂ Melting Experiment (Experiment 6).

Fig. 4.2. Fuel Residue from UO₂ Melting Experiments (Experiment 7).

Table 4.1. Material Released from the Fuel in UO₂
Melting Experiments (%)

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

Expt	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

Table 4.2. Material Released from High-Temperature Zone^a
of the Furnace in UO₂ Melting Experiments (%)

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

Expt	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	0.0773
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

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

materials in this zone were considered to be released from the fuel. Fission product release was generally greater in experiment 2, which had apparently reached a higher maximum temperature and which was irradiated unclad.

Table 4.2 gives the percentages of various fission products released from the high-temperature zone of the furnace. In experiment 2 strontium and barium were swept out of the furnace in the form of gaseous krypton and xenon by the helium during irradiation of the unclad fuel. The Ba^{140} , for example, was found in the charcoal trap, where it would be expected if it had been carried as Xe^{140} .

It is interesting to note from Tables 4.1 and 4.2 that the release of iodine and cesium from the fuel in experiment 7 was almost the same as the release in experiments in which the fuel melted and that the release of the nonvolatile fission products from the high-temperature zone was similar to the release in the other experiments even though the release of these fission products from the fuel in experiment 7 was considerably lower. It is considered significant that in these experiments much lower fractions of the nonvolatile fission products and slightly lower fractions of the volatile fission products were released from the high-temperature zone of the furnace than were released from the fuel.

One experiment has been conducted in which a miniature fuel element consisting of pyrolytic-carbon-coated uranium carbide in a graphite matrix was burned at temperatures up to approximately 1400°C . In this experiment fission and gamma heat in the irradiation position produced a sufficiently high temperature for the fuel to burn when air was substituted for helium. The sample was allowed to burn for 15 min, during which time approximately 59% of it was consumed. The remaining portion of the sample is shown in Fig. 4.3. Metallographic sections of coated uranium carbide particles which were subjected to various degrees of oxidation are shown in Fig. 4.4. These particles, which had fallen away from the burning fuel element, were recovered from the region around the holder.

Radiochemical analyses of the carbide experiment showed that only 38% of the uranium was liberated during the burning although 59% of the fuel element was consumed, indicating that the fuel was not homogeneous. Table 4.3 shows the release data from this experiment. In interpreting

Fig. 4.3. Fuel Residue from Uranium Carbide Burning Experiment.

Table 4.3. Material Released in First In-Pile Fuel Burning Experiment (%)

Sample length, 0.5 in.; diameter, 0.6 in. with 0.25-in.-diam axial hole; sample burning time, 15 min; air flow, 400 cc/min

	Sr ⁸⁹	Zr ⁹⁵	Ru ¹⁰⁶	I ¹³¹	Te ¹³²	Cs ¹³⁷	Ba ¹⁴⁰	Ce ¹⁴⁴	U
Release from fuel	13.8	0.681	48.2	28.8	42.7	51.3	14.3	9.05	2.12
Release from high-temperature zone ^a of furnace	1.20	0.478	40.7	26.8	35.1	36.9	0.499	6.24	0.369

^aHigh-temperature zone includes fuel material and thermal insulation; minimum temperature, 400°C.

the relatively low release from the fuel, it must be remembered that a significant fraction of the fission products was probably trapped inside those particles having undamaged pyrolytic carbon coating, especially those in the unburned portion. The release from the high-temperature zone

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stainless steel exit tube which carries gas and gas-borne activity from the furnace to the filters is being used as a diffusion channel to determine the diffusion coefficient for each of the fission products. This method of analysis, which has been described previously by Browning and Ackley,² interprets the observed distribution of the various fission products along the wall of the tube. The results of this type of analysis, as applied to the UO₂ experiments, are given here. Table 4.4 shows that there were apparently two particle-size groups in the millimicron range. One group was centered around an apparent size of 22 A. An average of 39% of the iodine carried out of the reactor furnace by the exit gas was carried by these particles; values in individual experiments ranged from 8 to 75%. No iodine having a diffusion coefficient corresponding to the vapor form was found. Some of the Cs, Sr, Te, and Ba were also found to be associated with this group of particles. The other group of particles was somewhat larger in diameter, 30 A, having a diffusion coefficient approximately half that of the smaller particles. Zirconium and uranium were associated with this group of particles, and ruthenium and cerium also appeared to be present in one experiment. However, ruthenium and cerium were usually carried on larger particles. For each isotope the remainder of the activity released from the furnace, exclusive of that tabulated, evidently was in the form of larger particles. The data from downstream parts of the exit-gas system are presently being analyzed to determine particle sizes. 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. This work should lead to the formulation of a model which explains the observed distributions and which may provide a basis for prediction of the behavior 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 conclusion, the results of both melting and burning in-pile experiments show that although large fractions of the fission products were

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²W. E. Browning, Jr., and R. D. Ackley, Nuclear Safety Program Semi-ann. Progr. Rept. June 30, 1962, ORNL-3319, p 44.

Table 4.4. Average Percentage of Each Isotope in Exit Gas Carried by Very Fine Particles

Effective Particle Diameter of Group, 22 A				Effective Particle Diameter of Group, 30 A			
I ¹³¹	39%	Cs ¹³⁷	15%	Zr ⁹⁵	3%	Ce ¹⁴⁴	13%
Sr ⁸⁹	5%	Ba ¹⁴⁰	13%	Ru ¹⁰⁶	10%	UO ₂	9%
Te ¹³²	3%						

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 A 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.

5. RELEASE OF FISSION PRODUCTS ON IN-PILE MELTING OF REACTOR FUELS UNDER TRANSIENT REACTOR CONDITIONS

G. W. Parker

R. A. Lorenz

In preparation for experiments on the release of fission products from UO₂ melted under transient reactor conditions, to be conducted in the TREAT (Transient Reactor Test Facility), an initial program proposal has been reviewed with TREAT management for the purpose of exchanging ideas before reaching a final design of the experimental assembly. In

general, the proposal appeared to satisfy the requirements outlined in the TREAT brochure.

The fuel piece and the experimental assembly, revised as a result of the above-mentioned discussions, have the appearance shown in Fig. 5.1. It was suggested that a pressure gage and valve be added as shown and that an overall assembly with graphite reflectors should be constructed using a nominal 4-in. stainless steel sheath tube for insertion into the lattice.

HAZARDS ANALYSIS

In preparation for the hazards analysis, leakage rates and thermal conduction have been studied in view of their importance to the safety of the experiment. The temperature profiles produced by heating the UO_2 capsule to 1300°C are shown in Fig. 5.2. Ten minutes was required to heat to 1300° at which time the external wall temperature of the autoclave was about 100°C . After heating 1 hr, with an internal atmosphere of 30 psi argon and an internal temperature of 1300°C , the external autoclave temperature reached 300°C . This indicates that there is a wide margin of safety in strength of the autoclave in case a high internal pressure should be encountered.

Leak testing has been performed both by cold hydrostatic pressure to 15,000 psi and by thermal cycling with 1000 lb helium internal pressure. Leakage was below the limit of helium detection (1×10^{-9} cc/sec) with no thermal cycling and with the standard 25-mil stainless steel gasket. A slight leakage of 1×10^{-8} cc/sec resulted on cycling from room temperature to 250°C . Gold plating the gaskets appears to reduce leakage on thermal cycling. For safety considerations it is significant that the expected maximum pressure of argon is about one-twentieth that of the helium used in these tests and that leakage of a significant amount of fission gas could occur only if the maximum observed leak rate was exceeded by a factor of more than 100. Thus a safety factor of 10^3 has been demonstrated by these tests.

Fig. 5.1. Experiment Assembly and Fuel Piece for Fission Product Release from UO_2 by Fast Transient Melting.

Fig. 5.2. Temperature Histogram in Components of TREAT Autoclave Assembly with a 1300°C Furnace Temperature.

GAS TRANSFER TO AN EVACUATED CONTAINER

The evacuated autoclave in the assembly is intended as a sampling device which is opened by an explosive valve following the transient. In order to assure that the filters will not be damaged by the pressure surge, it is proposed that a length of capillary tubing or other device be used to restrict the flow. The rate of pressure drop has been measured with a coiled 4-ft section of 20-mil-ID capillary tubing which will permit a safe transfer rate of ~1200 cc/min. An initial pressure of 60 psig completed the transfer in about 1 min.

MELTING SCHEDULE

A minimum amount of examination equipment (no hot cell facilities) will be required in order to conduct the initial series of tracer-level melting experiments. It is probable that this type of experiment can be conducted promptly, and materials are presently being obtained for this series. Hot-cell facilities are proposed for handling the preirradiated fuel specimens that will be used in later experiments. The designs of a dismantling assembly and shipping cask for use in connection with high-activity-level melting experiments are under study.

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6. FISSION PRODUCT TRANSPORT EVALUATIONS

C. J. Barton

M. E. Davis

G. W. Parker

PARTICLE-SIZE STUDIES

The previously described hot-cell installation, consisting of a box with Lucite walls and an aluminum frame, for use in studying particles produced in simulated reactor accident experiments, was completed. It will be installed in a hot cell for preliminary tests with unirradiated UO_2 , and possibly with slightly irradiated UO_2 , when the rf power equipment for melting the samples, described elsewhere,¹ is installed. This should be accomplished in the near future.

IDENTIFICATION OF CHEMICAL SPECIES

The apparatus designed to determine the heat of vaporization of radioactive species by a counting technique and the results of two exploratory experiments with radioiodine in this apparatus were described¹ earlier. This apparatus was modified by elimination of the side arm,

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¹C. J. Barton and G. W. Parker, Nuclear Safety Program Semiann. Progr. Rept. Aug. 17, 1962, ORNL-3319, p 39.

through which a steel rod was inserted to break the thin window of the iodine capsule. The modified version, made of quartz, had a quartz plunger, with an iron core, to break the window. A magnet held the plunger in position over the capsule. After adjustment of the helium flow and the negative pressure at the exit end of the apparatus, the seal was broken by de-energizing the magnet. With helium flowing through the apparatus at a constant rate and at a constant temperature, the rate of increase of the iodine content of the small charcoal trap near the top of the furnace tube was followed with a crystal detector connected to counting equipment which permitted both manual counts and continuous recording. The helium temperature was measured by an iron-constantan thermocouple junction just below the ampoule. Several experiments with this arrangement indicated that the transport of iodine from the ampoule to the charcoal trap was sufficiently rapid at temperatures around 100°C so that it was difficult to obtain equilibrium rate data. Another difficulty noted was that, after a fairly large fraction of the iodine had been transferred from the sample ampoule at some higher temperature, it was necessary to pass helium through the apparatus for some time before the rate of transfer of iodine dropped to its equilibrium value at or near room temperature. This seems to indicate that some of the iodine was adsorbed on the quartz wall of the furnace tube at a position between the sample ampoule and the trap, possibly because of poor heat distribution in this region.

The iodine transfer rate was reduced to a more manageable value by changing the sample ampoule from the thin-window type to one having a 1-mm-ID capillary tube, 4 cm long, through which the iodine had to diffuse before it reached the carrier-gas stream. The break seal on the side of the capillary was broken, after insertion of the ampoule in the apparatus, by removing the charcoal trap at the exit end of the furnace tube and inserting a long glass rod. The data obtained could be correlated by an Arrhenius-type plot, yielding heats of vaporization ranging from 13.0 to 15.0 kcal/mole (eliminating one experiment which produced badly scattered data). The average of six values obtained was 14.2 kcal, which agrees reasonably well with the published value² of 14.9 kcal for solid iodine.

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²F. R. Bichowsky, The Thermochemistry of Chemical Substances, Reinhold, New York, 1936.

It seems probable that this technique could be refined to diminish the scatter of data, but other factors, including the difficulty involved in extrapolating to the carrier-free situation and in applying the technique to release experiments, encouraged investigation of an alternate technique suggested by R. F. Newton. He postulated that the heat of vaporization might be determined by measuring, over a range of temperatures, the relative concentration of radioactive species in the vapor phase in an evacuated bulb which is in equilibrium with a solid or liquid phase in another bulb, to which it is connected by a capillary. The vapor-phase bulb is maintained at a higher temperature than the sample bulb in order to prevent deposition of solid or liquid material on the wall. The experimental arrangement is shown in Fig. 6.1.

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Fig. 6.1. Sealed-Tube Iodine Equilibration Apparatus.

Experiments to test this concept have been conducted with 1-cm-diam Pyrex bulbs connected by a capillary tube 10 cm in length and having an ID of about 1 mm. The experiments conducted to date have been with bulbs containing 0.20, 0.25, 0.40, and 4.1 mg of iodine carrier, along with about 0.1 mc of I^{131} . Typical data obtained are displayed in Fig. 6.2. The upper bulb, which was viewed by the crystal detector, was maintained at a fixed temperature in most of the experiments, but it is apparent that the slope of the curve obtained with a fixed temperature differential varies only slightly from that obtained with the fixed top bulb temperature. The data in Fig. 6.2 show that the surface effect noted in the iodine transport experiments is evidenced in the behavior of iodine at lower temperatures in the sealed Pyrex tubes and that the temperature at which deviation from the linear relation occurred cannot be readily correlated.

Fig. 6.2. Iodine Equilibration Data from Sealed-Tube Experiments.

with the amount of iodine present. Further tests will be required in order to determine whether the value for the heat of vaporization of solid iodine observed with the 0.4-mg sample (14.4 kcal) is reproducible and can be obtained with other amounts of iodine. The reason for the large variation in heat-of-vaporization values obtained to date is not clear, but it was recently observed that some sample ampoules contained a small amount of water which may help to account for the anomalous behavior. It may be significant that the 0.4-mg I_2 sample, which gave the best value for the heat of vaporization of iodine, showed no evidence of water contamination.

Since this technique could be more easily and safely applied to the study of released fission products than the transport method, further study is planned in an effort to find conditions which will yield usable data.

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7. CHARACTERIZATION AND CONTROL OF ACCIDENT-RELEASED FISSION PRODUCTS

The behavior of radioactive materials in gases depends markedly upon their form; consequently, an investigation of the dispersal or transport of such materials in gases, or the selection and design of systems for their removal from gases must take recognition of the distribution of the radioactivity among the different forms of material. A comprehensive analysis of the hazards associated with accident-released fission products requires determinations of (1) the distributions of the various radioactive nuclides among the different forms of materials in gases as a result of various accident conditions, (2) the behavior of these forms of materials in gases emerging from the reactor during and following an accident, and (3) the effectiveness of potentially useful gas-cleaning devices for the removal of the various forms of radioactive materials.

The work reported in this section is directed toward the problem of measuring the amounts of radioactive materials existing in various forms

in a gas stream and applying these measurement techniques to the determination of the efficiencies of gas-cleaning devices. Effective filters for particulate materials of approximately 0.1μ and larger in diameter and effective adsorbers are available for radioiodine vapor. Consequently, emphasis is being placed on other forms of materials which may be more difficult to remove from gases. It is expected that the measurement techniques developed in this work will also find application in other phases of the analysis of hazards resulting from the release of fission products.

DIFFUSIONAL CHARACTERIZATION OF MILLIMICRON RADIOACTIVE AEROSOLS
AND THEIR REMOVAL FROM REACTOR GASES

W. E. Browning, Jr.

R. D. Ackley

This work is a continuation and an extension of that reported in the previous issue.¹ An important mechanism in aerosol transport and deposition is that of diffusion, and, since the diameter of a particle is related to its diffusion coefficient, it is useful to characterize aerosols by their diffusion coefficients. A method for accomplishing this was developed in which diffusion coefficients for fine particles and for radioactive vapors such as iodine were determined by measuring the distribution of radioactivity on the walls of a channel previously exposed to gas carrying radioactive materials and flowing under laminar conditions.¹ Both cylindrical and rectangular geometries were employed; however, since the equation for a cylindrical geometry was not presented earlier and since cylindrical tubes are currently employed exclusively in this work, Eq. (1), corresponding to deposition of a given species in a cylindrical diffusion channel, obtained from the equation of Gormley and Kennedy,² is presented:

$$n_s = \frac{N_0 D}{Q} (9.4106 e^{-11.489 D Z / Q} + 6.8309 e^{-70.06 D Z / Q} + 5.8198 e^{-179.07 D Z / Q}), \quad (1)$$

¹W. E. Browning, Jr., and R. D. Ackley, Nuclear Safety Program Semiann. Progr. Rept. June 30, 1962, ORNL-3119, pp 44-50.

²P. G. Gormley and M. Kennedy, Proc. Roy. Irish Acad., sec A, 52, 163-69 (1949).

where

n_s = number of particles deposited per unit length,

N_0 = number entering the channel,

D = their diffusion coefficient,

Q = volumetric flow rate of carrier gas,

Z = distance from channel entrance.

For the case of a multiplicity of species, the deposition would be given by a summation of expressions of the form of Eq. (1). The diffusion-channel method was tested using radioiodine vapor, I^{131} -labeled 0.004- μ aluminum oxide particles, and I^{131} -labeled 0.25- μ tobacco smoke particles; the particle sizes and/or diffusion coefficients of these three materials were obtainable by independent means. In all three cases good agreement between theory and experiment was observed, indicating that this method is applicable over the range of interest.

This method of characterizing gas-borne radioactive materials is currently being applied to the problem of removing millimicron-diameter radioactive particles from air streams. Such particles may be too small in mass for efficient filtration by inertial impaction but sufficiently large, as compared with molecules of iodine vapor, that diffusion is too slow for efficient deposition.

Behavior of Millimicron Particles in Conventional Gas-Cleaning Systems

In the application of particle diffusion measurements, I^{131} -labeled aluminum oxide particles with a nominal diameter of 0.004 μ were adopted for standard usage. The particles were generated in air passing through an aluminum-wire spark gap, and this air stream was mixed with another carrying radioiodine vapor. After the mixture was aged and excess I_2 vapor was stripped out, the radioactive-particle-laden air stream was made to flow through the filter medium being tested, usually at a face velocity of 5 fpm. On either side of the filter medium, side streams under laminar flow conditions were drawn through the diffusion channels, which were silver-plated copper tubes 2 ft in length with an ID of 0.8 cm. Tests were made at room temperature and pressure. Changes were observed in the particle-size spectra due to a deliberate variation in aging time.

The results of a test of this type with a 0.75-in. bed of 6×16 mesh activated carbon were reported earlier. This test has been repeated, yielding substantially the same results as before. Both tests were conducted with a gas-flow velocity of 5 fpm, and the results can be summarized by stating that, whereas iodine vapor removal was essentially complete, the removal efficiency for 0.003- to 0.006- μ particles was observed to be only 75 to 90%, the higher efficiency being associated with the finer particles.

Two similar tests were performed with high-efficiency, low-pressure-drop, particulate filter media. The results of one test are shown in Fig. 7.1. This medium does not appear to be effective for removal of radioactivity in the form of iodine vapor or particles ranging up to 0.004 μ , since it exhibited an efficiency of only 10 to 75% in the two tests.

The fact that radioactive particles with diameters of approximately 0.003 μ are apparently produced on melting irradiated nuclear fuel is

Fig. 7.1. Removal of Very Fine Particles from Air by a High-Efficiency Particulate Filter.

discussed in Sec 4. This observation, together with the above-mentioned results obtained with charcoal adsorbers and high-efficiency filters, suggests that development of less-conventional gas-cleaning methods or improvement of existing methods may be required to ensure the safety of reactor containment systems. Accordingly, the work described next was undertaken.

Removal of Millimicron Particles by Less-Conventional Methods

In the first of these methods, the objective was to cause the radioactive particles to increase in size so that they could be removed by a high-efficiency filter. More specifically, the particles were intended to serve as condensation nuclei for steam. Figure 7.2 shows the apparatus employed. Cool air carrying particles was mixed with air saturated with

Fig. 7.2. Diffusion Apparatus Used for Investigating the Effect of Fog Formation on the Filtration of Very Fine Particles.

water vapor at about 95°C, resulting in fog formation. A condenser was used to remove excess water vapor in order to prevent interference with the use of the exit diffusion tube. The results of this study are summarized in Fig. 7.3; the left side of the figure corresponds to a control experiment. Comparison of the data for filtration with and without prior fog formation indicates removal efficiencies of 99% vs 85% for 0.002- μ

Fig. 7.3. Effect of Fog Formation on the Filtration of Very Fine Particles.

particles and 85% vs 35% for 0.007- μ particles. These results are encouraging, but the improvement in removal efficiencies is not high enough to provide the basis for a final design. Optimization of this method might provide a partial answer to the problem of small-particle containment; however, it appeared desirable to first explore some other possible methods.

If air carrying millimicron particles is used to generate foam or small bubbles, the particles might diffuse to the air-liquid interface and be retained. The use of foam for removal of iodine and $\geq 0.074\text{-}\mu$ particles from air has previously been reported by Silverman and co-workers at Harvard.³ Figure 7.4 shows the apparatus used to investigate the removal efficiency of foam when air carrying radioactive aluminum oxide particles was the dispersed or internal phase. The dispersion medium, or

³L. Silverman, M. Corn, and F. Stein, "Diffusion Board Containment Concepts and Foam Encapsulation Studies," Proceedings of the Seventh AEC Air Cleaning Conference, October 10-12, 1961, TID-7627, pp 390-405 (March 1962).

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Fig. 7.4. Diffusion Apparatus for Investigating Efficiency of Foam for Removal of Very Fine Particles from Air Streams.

external phase, was an aqueous solution of certain organic liquids formulated to produce a given height of foam under the operating conditions. In an actual application the air in a reactor vessel might be recirculated through a pool of water containing a suitable foaming agent. Three tests were performed: one with a 1-ft and one with a 3.5-ft foam column and one with a 5-in. column or pure water for a control. The control test showed negligible removal of the nominally 0.007- μ component but about 95% removal of a composite component believed to include a mixture of iodine vapor and < 0.002- μ particles. The results obtained with a 3.5-ft column are shown in Fig. 7.5, which indicates a removal efficiency of about 85% for the 0.007- μ component. Rather surprisingly, the results for the 1-ft foam column were not appreciably different from those obtained with the 3.5-ft column. Plans are to investigate this aspect further and, also, to test other promising methods for the removal of very fine particles from gases.

The various results pertaining to removal methods for ultrafine particles are summarized in Table 7.1. The data show that millimicron particles are capable of carrying radioactive materials through conventional gas-cleaning devices. Some progress has been made in developing more efficient methods for the removal of such materials from gases, and these studies are being continued.

Fig. 7.5. Removal of Ultrafine Particles from an Air Stream by a Foam Column.

MEASUREMENT OF CHARACTERISTICS OF RADIOACTIVE AEROSOLS USING FIBROUS FILTERS

W. E. Browning, Jr.

M. D. Silverman

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 is being developed for measuring aerosols in terms of their response to these processes by determining their distribution as a function of fibrous-filter depth under carefully controlled conditions. It is anticipated that this work will also extend the theory of the performance of fibrous filters to conditions for which little information is now available.

The measurement of the distribution of aerosol materials with depth in ordinary paper filters is made difficult by the thinness and the complexity of the medium. Sisefsky, in a short note to *Nature*,⁴ described

⁴J. Sisefsky, *Nature* 182, 1437 (1958).

Table 7.1. Observed Efficiencies of Various Methods for Removal of Ultrafine Particles and/or Radioiodine Vapor from Air

Method	Identity or Nominal Size of Component	Removal Efficiency (%)
High-efficiency filter	I ₂ and/or < 0.002 μ	75
	0.002 μ	< 10
	0.004 μ	~ 50
Activated-carbon bed	I ₂	> 99
	0.005 μ	75-90
Fog chamber and filter	Without fog formation	
	0.002 μ	85
	0.007 μ	35
	With fog formation	
	0.002 μ	99
	0.007 μ	85
Foam column		
	5-in. water column (no foam)	
	I ₂ and/or < 0.002 μ	95
	0.007 μ	~ 10
	1-ft foam column	
	I ₂ and/or < 0.002 μ	98
3.5-ft foam column	0.007 μ	85
	I ₂ and/or < 0.002 μ	99
	0.007 μ	85

a method for determining particle penetration depths. He passed an aerosol containing radioactive solid particles 0 to 2 μ in diameter through a thin glass-fiber filter and obtained approximate information on the relative sizes and amounts present by peeling off successive layers of the filter using pressure-sensitive tape. Interpretation of these data in the literature is impeded by poor depth resolution and by the large range of diameters of the fibers employed.

Theoretical calculations performed to determine the effects of various conditions on the interaction of aerosols with fibers showed that fiber diameter has a very strong effect. The wide variation in diameter of the fibers employed in commercial fibrous filters would necessitate the use of approximations and averages in handling the data obtained from

filtration experiments involving these media. This difficulty was overcome by obtaining a commercial synthetic fiber, Dacron polyester staple, of unusually uniform size (99%, $11.3 \pm 0.8 \mu$) from Du Pont. The fiber was carded into a web and rolled to give a product which had the appearance and feel of filter paper. A number of 1-1/2-in.-diam disks of this material, made by using a metal punch, were placed between metal washers and enclosed in a Teflon holder to give a compact filtering device. Entrance and exit channels, of sufficient length to avoid gas turbulence, contained O rings which fitted against the end washers to provide a seal within the Teflon holder. This arrangement facilitated separation of the fiber bed into discrete layers for radioassay after exposure to the aerosol.

A radioactive aerosol containing Zn^{65} has been developed for initial tests of this method of characterizing aerosols. Specimens of 0.010-in.-thick zinc foil were irradiated in the LITR reactor. A spark generated by a Tesla coil was passed across the gap between two pieces of the foil, placed approximately 1/16 in. apart. A stream of air which passed over the electrodes carried the aerosol through the system containing the series of filters. A diagram of the apparatus is given in Fig. 7.6. An electron micrograph (Fig. 7.7) of samples of this aerosol collected on a membrane filter shows that it consisted of particles with diameters in the range 40 to 300 Å.

Fig. 7.6. Apparatus for Characterization of Aerosols Using Fibrous Filters.

Fig. 7.7. Electron Micrograph of a Zn⁶⁵ Aerosol.

The data obtained for the first series of experiments are presented in Fig. 7.8. Although preliminary in nature, they indicate that the techniques used are satisfactory and should provide useful information on aerosol character and on filtration mechanisms. The slope of the distribution curve on a semilog plot equals the coefficient of interaction between aerosol and fibers and is composed of terms corresponding to each of the interaction processes. This coefficient is characteristic of the aerosol species. Data are shown for four different gas velocities. It is evident that at 0.7 and 1.7 fpm, the deposit of activity is reduced

Fig. 7.8. Distribution of Zn^{65} Aerosol Through Dacron Fiber Filters.

by a factor of 10^4 , or 99.99%, whereas at 2.8 and 9.8 fpm, reduction is not as great, 10^3 or 99.9%, over a filter depth which contains 10^5 cm of fiber length per square centimeter of filter area. At this time it is uncertain whether the tails of the curves are meaningful, since the lowest counting rates were near background. Future experiments will be performed with more highly irradiated specimens which should provide higher counting rates and, it is hoped, more data in this area.

All the flow rates are within the viscous or laminar flow region. At the lowest linear velocity (~ 0.7 fpm), where diffusion would be controlling, collection efficiency is increased by decreased velocity as expected. At the higher flow rates, where the primary mechanism of filtration is by interception for particles of this size, the efficiency appears to be independent of velocity, in accord with theory. This investigation is being continued to provide a more specific identification of the mechanisms which produce the distribution patterns observed. Interpretation of distributions in terms of aerosol characteristics will then be possible.

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PART III CONTAINMENT ENGINEERING

REACTOR DIVISION

H. G. MacPherson, Division Director

W. B. Cottrell, Coordinator

8. NUCLEAR SAFETY PILOT PLANT

M. H. Fontana

L. F. Parsly

The purpose and design of the Nuclear Safety Pilot Plant were described in the previous semiannual report.¹ Work during the past six months on the NSPP has been principally in the preparation of detailed drawings and specifications for procurement and fabrication of components. Inasmuch as installation of the NSPP components into the designated cell was not scheduled to begin until early in 1963, arrangements were made with the Harvard Air Cleaning Laboratory and the AEC to conduct a special experiment in the cell during December. This experiment will determine the effectiveness of foam in trapping and holding fission products released to the atmosphere.

STATUS OF DESIGN AND CONSTRUCTION OF NSPP

Only two changes of any significance have been made in the design concept during the period reported. The particulate filters and iodine removal beds in cell B were eliminated when it was determined that the existing stack filters in Building 7500 were adequate. Also, since a furnace capable of handling fuel elements 6 in. long should be adequate during at least the first 2 yr of operation, the design capability of the furnace was reduced to this figure. It is anticipated that experience

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

in melting short fuel elements will lead to design improvements by the time a furnace capable of melting 30-in. fuel elements is needed.

During the report period a preliminary design of the furnace for handling 30-in.-long fuel elements was completed, and the design of the furnace for handling 6-in.-long fuel elements has been completed and issued for construction. A preliminary operating procedure was prepared as a basis for design of shielded carriers, cell shielding, and remote tooling. Design studies of the tools, carriers, and shielding are in progress.

The model containment vessel has been fabricated, and delivery is expected before the first of the year. Virtually all components have been released for fabrication, and a few small components have been received. Installation of equipment in cell B, Building 7500, is scheduled to start in March 1963.

FISSION PRODUCT TRAPPING BY FOAM

The Oak Ridge National Laboratory is collaborating with the Harvard Air Cleaning Laboratory on an intermediate-size test of the encapsulation of released fission products within foam.² This scheme was developed by Silverman et al. at Harvard, where it was tested in laboratory-scale facilities with tracer amounts of I^{131} . The test at Oak Ridge will be performed with up to 10 curies of I^{131} and similar quantities of mixed fission products in the 6000 ft³ cell B. This is the same cell which will be the site of the Nuclear Safety Pilot Plant. However, it is expected that the foam tests will be completed and the cell cleaned up by the first of January, several weeks before installation of the NSPP components is scheduled to commence.

The principle behind the use of foam in the containment of activity released from a nuclear incident is that the foam will encapsulate the released fission products.

Contaminated air is drawn through a foam generator and returned to the cell volume as foam until the cell is full of foam. The subdivision of the cell to millions of smaller volumes increases the surface available

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²Leslie Silverman, "Foam and Diffusion Board Approaches to Containment of Reactor Releases," presented at the Third Conference on Nuclear Reactor Chemistry, Gatlinburg, Tenn., Oct. 9-11, 1962.

for deposition of the fission products and decreases the distance over which the nuclides must travel by diffusion to reach a surface. When the foam decays, as it does with a 12-hr half-life, the resulting liquid should contain most of the contamination previously airborne in the cell.

A schematic layout of the cell for the foam tests is shown in Fig. 8.1. The cell will be sealed by plastic and masking tape and maintained at a negative pressure by the stack fans. Any flow through the stack system will pass through a particulate filter and a charcoal filter before leaving the stack. Desired pressure differential between the operating floor and the cell will be maintained by remote manipulation of a damper in a parallel line to the stack.

Sampling will be performed by drawing cell air through heated aluminum lines to a charcoal sample collector, by means of a vacuum pump which will discharge back to the cell. The sample lines will be heated to approximately 160°F to minimize excessive iodine deposition before the cell air reaches the charcoal collector. The lines will sample three positions along the vertical midpoint of the cell.

The radioisotope source will be I^{131} , obtained as palladium iodide in a glass capsule. The capsule will be lowered into the furnace in an aluminum bucket by remote handling and will be broken remotely. The iodine will be released to the cell by decomposition of the PdI_2 .

The effects of the foam will be determined by comparison of tests in which foam is used with tests in which no countermeasures are used. After each test the foam will be collapsed by means of a water spray and the contaminated liquid transferred to a 12,000-gal storage tank. Sampling may be performed at this point to determine the activity retained in the solution.

Cell B has been prepared for the experiment, and shakedown tests are being performed. The proposed experiment and its associated hazards have been described and analyzed,³ and approval was given to proceed. Results of the test will be summarized in the next semiannual report.

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³J. R. Buchanan and M. H. Fontana, Foam Suppression Tests in HRT "B" Cell, ORNL CF-62-9-36 (Sept. 9, 1962).

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Fig. 8.1. Schematic Layout of Cell for Foam Tests.

9. REACTOR CONTAINMENT HANDBOOK

T. H. Row

The Reactor Containment Handbook being prepared by ORNL will contain information useful in the design of reactor containment systems. For example, information will be provided on suggested design procedures, data, experience, credible accidents, and process descriptions.

Four chapters of the handbook are to be prepared under subcontract by Bechtel Corporation. The contract was signed October 25, 1962, and Chapters 7, 9, 10, and 11 are scheduled for completion in draft form by March 1, 1963.

The handbook outline given previously remains basically the same.¹ The sections and their status are given below.

1. Introduction (> 50% complete)
2. Codes, Criteria, and Regulations (completed, to be issued as an ORNL CF memorandum)
3. Radioactivity - Generation and Transport (< 50% complete)
4. Energy Sources (completed, to be issued as ORNL CF memorandum)
5. Analytical Techniques (< 50% complete)
6. Credible Accidents (< 25% complete)
7. Containment Proof Tests (Bechtel Corporation, literature survey proceeding)
8. Specific Containment Systems (< 25% complete)
9. Materials Considerations for the Use in Analysis (Bechtel Corporation, collection of basic materials data proceeding)
10. Construction Details and Accessories (Bechtel Corporation, literature survey and assignment of subsections in process)
11. Economics (Bechtel Corporation, literature survey, detailed outline, and writing of draft material started)
12. Containment Research (not started)

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¹C. L. Whitmarsh, Nuclear Safety Program Semiann. Progr. Rept. June 30, 1962, ORNL-3319, pp 63-65.

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PART IV RADIOCHEMICAL PLANTS

CHEMICAL TECHNOLOGY DIVISION

F. L. Culler, Division Director

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10. RADIOCHEMICAL-PLANT SAFETY STUDIES

C. E. Guthrie

E. D. Arnold

J. P. Nichols

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 primary effort during this period has been a hazards analysis on a hypothetical 1-ton/day spent-fuel processing plant. The accidents considered were (1) an explosion in the high-level waste evaporator and (2) a criticality accident in the plutonium product evaporator. Briefly, the results of the analysis are as follows: (1) The consequences of the accidents are not serious as long as there is good primary and secondary containment and the cell off-gas filters remain intact. (2) Failure of

either the primary containment or the secondary containment would result in the release of contamination to the surroundings, but the area of contamination would not be large. (3) Primary containment is more important than secondary containment, mostly because of possible lethal exposure to personnel in the building if primary containment fails. (4) An accident involving failure of both primary and secondary containment would be extremely serious and would contaminate an extended area ($> 10^5 \text{ m}^2$) under average meteorological conditions. (5) Under the worst meteorological conditions (washout) failure of the filters is just as serious as failure of both primary and secondary containment and could result in contamination of up to 10^6 m^2 .

Aerosol concentrations produced in various accidents and their properties were basic assumptions required for the study. Before radiochemical-plant hazards can be evaluated with more certainty, a study must be made of the amount of material that can be dispersed and of the properties of aerosols in flowing through ducts and cracks. In addition, in view of ORNL experience with absolute filters in chemical-plant service, it is evident that there is a high probability that the filters would not be intact in case of an accident (either as a result of the accident or of degradation during normal operation). The development of a more reliable chemical-resistant filter is, therefore, vital to the safety of a radiochemical plant. Work on radiochemical-plant safety will concentrate on aerosol production and dispersion studies and filter development.

RADIOCHEMICAL-PLANT HAZARDS EVALUATION

Types of Dispersive Accidents

The most serious accidents that may credibly occur in large radiochemical facilities are chemical and nuclear explosions which rupture vessels filled with radioactive process solutions or solids. It is our current belief that the maximum credible accident in a radiochemical plant would correspond in gas production and blast effects to an explosion of 3 lb of TNT. (Three pounds of TNT liberates approximately 5700 Btu of energy, generates approximately 100 ft^3 of hot gases, and creates a shock

wave that has a pressure of approximately 800 lb/ft² and an energy of approximately 230 ft-lb/ft² at a distance of 15 ft.) Thick concrete cells of the type used in ORNL radiochemical facilities can withstand such explosive effects without rupture.

Effects of Dispersive Accidents

In the maximum chemical explosion the radioactive liquid would be sprayed through the cell, forming an aerosol of radioactive material in the cell air. A fraction of this aerosol would reach the environment through the vessel off-gas system and cell ventilation system and through successive leaks from the cell and from the building. The maximum nuclear burst would, in addition, disperse new gaseous fission products and expose operating personnel to prompt neutron and gamma radiation through the shield. The maximum integrated dose through a 5-ft-thick concrete wall would be less than 1 rem if personnel evacuate the facility within 2 min.

Bases of Calculations

The bases used for the hazard analyses are shown in Table 10.1. The physical constants of the facility were chosen to be typical of those for a 1-ton/day spent-fuel processing plant meeting ORNL containment standards.

The activity dispersed in the cell was assumed¹ to be 1 cell volume of an aerosol with a concentration of 10 mg/m³. This assumption is basic to the calculations since the total dose and fallout are directly proportional to the value assumed. It is quite possible that a higher concentration would be stable as far as the top of the stack or that a cell fire could yield a much higher volume of aerosol. This point requires further investigation.

The contamination limits for Sr⁹⁰ were based on the level that would impose restrictions on agriculture.² The Pu²³⁹ level was based on a re-

¹E. D. Arnold, A. T. Gresky, and J. P. Nichols, "The Evaluation of Radioactive Releases from Chemical Plants," pp 222-41 in Seventh AEC Air Cleaning Conference held at Brookhaven National Laboratory, Oct. 10-12, 1961, TID-7627 (March 1962).

²"Effects of Fission Product Release on Humans and Land Use," p 40 in Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants, WASH-740 (March 1957).

Table 10.1. Bases of Calculations

Processing Plant

1. Capacity: 1 metric ton of uranium per day
2. Fuel: Yankee Reactor, 8400 Mwd/metric ton, 120 days' cooling
3. Waste-evaporator fission-product concentration, 1080 curies/liter
4. Maximum plutonium concentration, 100 g/liter
5. Cell volume, 8000 ft³
6. Secondary containment volume, 3×10^5 ft³
7. Ventilation-system transit times: vessel off-gas, 150 sec; cell off-gas, 30 sec
8. Stack dilution factor, 1.6×10^{-5} sec/m³
9. Cell ventilation rate, 800 cfm
10. Primary containment pressure, -1 in. H₂O; leak rate, 0.01 cell volume per minute at -2 in. H₂O
11. Secondary containment pressure, 0 to -0.3 in. H₂O in 20 sec; leak rate, 6×10^{-3} building volume per minute at 0.3 in. H₂O

Activity Release in Cell

1. Cell aerosol concentration, 10 mg/m³
2. Waste-evaporator explosion: aerosol concentration, 0.129 curie/m³; total suspended, 29.2 curies
3. Plutonium criticality incident: plutonium aerosol concentration, 5.12×10^{-5} curie/m³; total plutonium suspended, 1.17×10^{-2} curie; number of fissions, 10^{18}

Contamination Limits for Cleanup

1. Pu²³⁹, 1.5×10^{-9} curie/m²
2. Sr⁹⁰, 7.5×10^{-8} curie/m²

suspension factor³ of 4×10^{-10} ($\mu\text{c}/\text{cm}^3$)/($\mu\text{c}/\text{m}^2$) and (from ref 4) an $\text{MPC}_a = 6 \times 10^{-13}$ $\mu\text{c}/\text{cc}$. Contamination levels were not calculated for I¹³¹ because of the short duration of restrictions due to the 8-day half-life.

³J. W. Healy and J. J. Fuquay, pp 427-36 in Progress in Nuclear Energy, Series XII, Health Physics (ed. by W. G. Marley and K. Z. Morgan), vol 1, Pergamon, London (1959).

⁴"Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and in Water for Occupational Exposure," National Bureau of Standards Handbook 69 (June 1959).

Methods of Calculation

The methods of calculation used for estimating the release, dose, and deposition are those developed for hazards analysis of ORNL facilities and are reported in some detail in the literature.^{1,5,6}

Two meteorological conditions were used in the evaluation of fallout resulting from the accident. The first is typical of average weather conditions and is the same as has been used in the previous studies. The ground concentration as a function of distance and type of release for these conditions is shown in Fig. 10.1.

⁵R. E. Tomlinson, Nuclear Safety 3(1), 56 (1961).

⁶E. D. Arnold and J. P. Nichols, Hazards Analysis of Fuel Handling Facilities, ORNL TM-346 (1962).

Fig. 10.1. Fallout Isopleths as a Function of Distance Downwind, Wind Velocity, and Height of Release.

The second meteorological condition is one of washout and is typical of the worst possible conditions with respect to fallout. The maximum credible enclosed area and the corresponding maximum downwind distance for a washout deposition isopleth were calculated with the assistance of the U.S. Weather Bureau Office.⁷ Credible conditions are defined as those representing practical, consistent weather conditions and particulate deposition velocities ≥ 0.1 cm/sec. The maximum area enclosed within a specific isopleth varies with weather conditions and with deposition velocity.

⁷Private communication with W. M. Culkowski, U.S. Weather Bureau, Oak Ridge, Tenn.

Fig. 10.2. Enclosed Area and Downwind Distance as a Function of Deposition Isopleth, Washout Conditions.

This is illustrated by two washout conditions that are plotted in Fig. 10.2. Heavy rain during daytime conditions causes largest deposition near the source, while a daytime light rain causes largest deposition distant from the source. Maximum deposition very near to the source is caused by deposition of large particles with high settling velocities.

Results and Conclusions

The possible personnel exposures, both in the building and downwind, and the contamination of surrounding areas were calculated for the two accidents for cases where primary and secondary containment held and the filters were 99.97%, 98.6%, and 0% efficient and also where the filters were 99.97% efficient but the primary containment alone failed, the secondary containment alone failed, and both containments failed. The results are summarized in Table 10.2. Significant personnel exposures and areas of contamination are underlined.

The maximum credible accidents in radiochemical facilities which have adequate primary and secondary containment and 99.97% efficient absolute filters systems (column 2, Table 10.2) would result in acceptably low personnel exposure and downwind ground contamination even under the most adverse meteorological conditions.

No serious contamination of surrounding areas or radiation exposure to personnel outside the building would result from rupture of either of the two containment systems (columns 5 and 6, Table 10.2). Building personnel would receive a significant (possibly lethal) exposure only in case of a primary-containment rupture.

Contamination of large areas could be expected in the case of a rupture of both primary and secondary containment. Significant personnel exposures at distances greater than 10 m would be expected only in case of a criticality accident and the rupture of both containment systems.

The maximum dose to personnel from I_2 released from the stack in the case of a criticality accident is sufficiently low (~55 millirem) that I_2 absorbers in the off-gas system would not be justified.

Table 10.2. Summary of Radiological and Fallout Hazards from Maximum Credible Accidents
in 1 Metric Ton/Day Spent-Fuel Processing Plant

Significant exposures and areas of contamination are underlined

	Containment Intact			Filters 99.97% Efficient		
	Filters 99.97% Efficient	Filters 98.6% Efficient	Filters Rupture	Secondary- Containment Rupture	Primary- Containment Rupture	Both Containments Rupture
Waste-Evaporator Explosion						
Aerosol Release						
From stack, curies	9×10^{-3}	0.41	29.2	9×10^{-3}	9×10^{-3}	9×10^{-3}
Into building, curies	1.6×10^{-2}	1.6×10^{-2}	1.6×10^{-2}	1.6×10^{-2}	28.8	28.8
From building, curies	3.2×10^{-5}	3.2×10^{-5}	3.2×10^{-5}	1.6×10^{-2}	5.7×10^{-2}	28.8
Consequences						
2-min dose in building, rem	3.46×10^{-2}	3.46×10^{-2}	3.46×10^{-2}	3.46×10^{-2}	63	63
Maximum downwind (10 m) dose, rem	4.0×10^{-6}	1×10^{-3}	7.2×10^{-2}	4.2×10^{-4}	1.5×10^{-3}	7.6×10^{-1}
Area of Sr^{90} contamination, m^2						
Washout, maximum	600	2×10^4	10^6	1000	3000	10^6
Fallout	0	0	0	40	60	10^5
Nuclear Excursion in Plutonium Extraction System						
Plutonium Aerosol Release						
From stack, curies	3.5×10^{-6}	1.6×10^{-4}	1.17×10^{-2}	3.5×10^{-6}	3.5×10^{-6}	3.5×10^{-6}
To building, curies	6.4×10^{-6}	6.4×10^{-6}	6.4×10^{-6}	6.4×10^{-6}	1.17×10^{-2}	1.17×10^{-2}
From building, curies	1.3×10^{-9}	1.3×10^{-9}	1.3×10^{-9}	6.4×10^{-6}	2.3×10^{-5}	1.17×10^{-2}
Consequences						
Dose from plutonium aerosol, rem						
Downwind, maximum	2.8×10^{-5}	1×10^{-3}	9.4×10^{-2}	5.7×10^{-4}	2×10^{-3}	1.03
2-min dose in building	4.6×10^{-2}	4.6×10^{-2}	4.6×10^{-2}	4.6×10^{-2}	84	84
Dose from gaseous fission products, rem						
Downwind, maximum	5.5×10^{-2}	5.5×10^{-2}	5.5×10^{-2}	5.5×10^{-1}	2	1000
2-min dose in building	1.5	1.5	1.5	1.5	2700	2700
Area of Pu^{239} contamination, m^2						
Washout, maximum	200	10^4	10^6	500	1000	10^6
Fallout	0	0	0	30	80	10^5

Because the activity is released from a stack, rather than at ground level, the consequences of an accident when there is a low filter efficiency or a filter rupture is strongly dependent on meteorological conditions. Under favorable meteorological conditions the material will be sufficiently dispersed so that fallout will not result in contamination levels high enough to impose restrictions. Under the most unfavorable meteorological conditions (washout) a stack release is just as serious as a ground release. During washout conditions a filter rupture could be disastrous (Table 10.2, column 4). Even in cases where the filter efficiency is 98.6%, contamination of large areas could result during washout conditions.

For some time ORNL has had a program of in-place testing of absolute filter systems.⁸ The results of initial in-place efficiency tests on systems under this program are shown in Table 10.3.⁹ In these tests and other

⁸R. W. Schneider, "Procedures for Testing High Performance Filters at ORNL, pp 187-214 in Seventh Air Cleaning Conference held at Brookhaven National Laboratory, Oct. 10-12, 1961, TID-7627 (March 1962).

⁹R. W. Schneider, "In-Place Testing of High Efficiency Filters," Nuclear Safety 4(3), sec IV (1963).

Table 10.3. Results of Initial Tests on ORNL Filter Systems

	Number of Systems		
	Type A ^(a)	Type B ^(a)	Type C ^(a)
Efficiency of 99.97% or higher	129	122	5
Efficiency of less than 99.97%	10	55	23
Total	139	177	28
Percent of total with efficiency of 99.97% or higher on initial test	93	69	18

- (a) Type A: One- and two-filter systems primarily of the chemical laboratory exhaust-hood type, in service 4 months or less.
 Type B: Same as type A, except in service 14 months or less.
 Type C: Larger multifilter installations of various types in service for various periods of time.

inspections there have been at least two instances where the filters had essentially zero efficiency. It is apparent from these results that AEC absolute filters in chemical-plant service may deteriorate rapidly and cannot be counted on for 99.97% efficiency (or even to be intact) in case of an accident. In view of the possible serious consequences as a result of a filter failure in a radiochemical-plant accident, it is imperative that an improved filter system be developed to assure radiochemical-plant safety.

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