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NUCLEAR SAFETY PROGRAM
SEMIANNUAL PROGRESS REPORT
FOR PERIOD ENDING JUNE 30, 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 June 30, 1962

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SUMMARY

1. INTRODUCTION

This is the first of a series of semiannual reports describing the Nuclear Safety Research and Development Program which is carried out at the Oak Ridge National Laboratory. Initial steps were taken at ORNL in 1955 to study the mechanisms and phenomenology of reactor accidents, and the scope of the program was enlarged in July of 1961. Previous work in this program is referenced in a bibliography.

2. REACTOR FUEL STORAGE AND HANDLING STUDIES

Critical and safe conditions for Rover fuel were defined under conditions characteristic of both its fabrication and storage. Since this work is classified, it is described in a separate memorandum, Critical Mass Studies, Part XII: Rover Reactor Fuel Elements, ORNL TM-264 (to be published), by E. B. Johnson and J. K. Fox.

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

The facilities of the Bulk Shielding Reactor I are being used to determine the reactivity effects of small fuel displacements in a pool-type reactor. Special fuel elements and loadings in a 25-element array have made possible the measurement of reactivity effects which would result from some geometrical changes. The types of changes used in the experiments were designed to test the validity of calculation methods for computing the types of perturbations that might result from a reactor excursion. The experiments indicate, thus far, that negative reactivities result from all perturbations that decrease the homogeneity of the core, except when the perturbations add fuel to high-importance regions. Calculations with the MODRIC and PDQ code appear to be inadequate to cope with the perturbations represented by the experiments.

4. RELEASE OF FISSION PRODUCTS ON OUT-OF-PILE MELTING REACTOR FUELS

The effect of irradiation level on fission-product releases from irradiated UO_2 when heated in air to 1200°C was studied under out-of-pile conditions. In general, the release rate was found to increase with

increasing irradiation levels. When specimens were heated in purified helium rather than air, higher release rates were also observed at the higher irradiation levels, with the greatest effect being observed in the case of cesium and the least in the case of ruthenium. When UO_2 specimens were melted, the high fission-product releases which were observed largely obscured the effect of burnup. The particles evolved from the UO_2 melt ranged from 0.01 to 0.1 μ in diameter, and the fission products were removed from the gas phase in an amount proportional to the amount of uranium oxide deposited on the equipment walls. In future studies, larger scale meltdowns of UO_2 clusters which are cored with resistance heated tungsten rods will be carried out.

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

In order to measure fission-product releases under more realistic conditions than out-of-pile experiments, meltdown of stainless steel clad UO_2 specimens was carried out in the Oak Ridge Research Reactor. In the five meltdowns which were performed, it was established that controlled melting of UO_2 could be achieved in the reactor and that, in general, the fission-product release values were consistent with those observed in out-of-pile experiments. The physical state of the specimens after melting varied markedly with the fission heating to which they were subjected; for example, in the case of low heat generation, the outer shell of the UO_2 failed to melt but a center void was produced. At higher heat generations, the fuel either melted completely or a needle-like deposit, presumably resulting from UO_2 vaporization, formed on the surface of the specimen. In these experiments iodine and the rare gases, krypton and xenon, are released to the greatest extent. The Ba^{140} and Sr^{89} behaved similarly to each other and Ce^{144} was released to a lesser degree. Although a large fraction of the fission products escaped from the UO_2 melt, they were almost entirely retained inside the high temperature zone of the furnace.

6. FISSION PRODUCT TRANSPORT EVALUATIONS

A study has been initiated to obtain information needed to attain a better understanding of the many factors which affect the transport and deposition of particles and gases which would be released in a fuel meltdown accident. An aluminum and Lucite fallout chamber is being built to make preliminary studies on the size distribution of gas-borne particles. A stainless steel fallout chamber has been designed for the study of particles released on melting comparatively large quantities (up to 80 g) of highly irradiated UO_2 . Two exploratory experiments have been made to give information on the behavior of iodine, adsorbed on charcoal, as a function of temperature, gas flow rate, and gas composition. It was found that a helium sweep gas, at room temperature, would remove a considerable amount of I^{131} adsorbed on charcoal. Increasing the temperature increased the transfer rate.

7. CHARACTERIZATION AND CONTROL OF ACCIDENT-RELEASED FISSION PRODUCTS

An investigation of the transport and deposition of radioactive aerosols was initiated to provide information for aiding in the control of the radioactivity which may be released in this form during a reactor accident. Thus far, the study has been mainly concerned with diffusion, both as an important mechanism in aerosol behavior and as a basis for the determination of particle-size distributions. Air containing radioactive iodine vapor or finely divided particulate matter labeled with I^{131} is passed through cylindrical or rectangular channels, and the resulting longitudinal activity profiles are determined by means of a gamma spectrometer. The deposition profile for an aerosol or vapor is governed by the diffusion coefficient, which, in turn, depends on particle diameter or molecular properties in the case of a vapor. The initial results obtained appear promising with regard to aerosol characterization, since iodine-labeled particles approximately 0.25 and 0.004 μ in diameter, as well as iodine vapor, behaved in accordance with theoretical equations adapted from the literature. The character of radioactive aerosols may thus be determined by deposition measurements, making possible more accurate predictions of the behavior of radioactive materials in gases in a reactor accident. Characterization of aerosols is also being attempted by the use of fibrous filters.

8. NUCLEAR SAFETY PILOT PLANT

The Nuclear Safety Pilot Plant is an experimental facility designed to study the release, transport, and removal of fission products in a model containment vessel. The facility will consist of a plasma torch heat source to melt fuel material; a model containment vessel 10 ft in diam and 15 ft high; sampling, off-gas, and decontaminating equipment; and associated instrumentation. The facility will be completely contained in a 12 x 25 x 23 ft cell in the 7500 building which will permit experimentation with more than 1000 curies. Process and instrumentation flowsheets have been prepared, the torch purchased, and the containment vessel let for bids. Detail lay-outs and furnace development are currently under way and it is expected that the facility will be completed in the spring of 1963 and in operation within a year.

9. REACTOR CONTAINMENT HANDBOOK

A Reactor Containment Handbook now being prepared by ORNL is scheduled for completion in 1963. A generalized outline of the Handbook is given to indicate the scope of the material to be covered. Preparation of draft material is approximately 15% complete.

10. RADIOCHEMICAL PLANT SAFETY STUDIES

Before a large radiochemical economy develops it is vital that the hazards associated with the industry be thoroughly evaluated. In order to accomplish this: (1) the major radiochemical plant accidents and their consequences to the environment must be determined; (2) the significant variables must be studied; and (3) means of combating the accidents or ameliorating their consequences must be developed. A radiochemical plant hazard analysis has been started covering fuel reprocessing and isotope source preparation plants. The current absolute filters are obviously a weak link in radiochemical plant safety. Preliminary investigation indicates that graphite fibers may be used to construct a much more reliable filter for radiochemical plant service.

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1. INTRODUCTION

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

In the operation of nuclear reactors, sizable quantities of dangerous fission products are generated in the fuel. In order to assure that the public is adequately protected, reactors are built inside containment vessels and are constructed in remote locations. Both of these precautions are costly, and it is generally conceded that the economics of nuclear power could be improved if the public safety could be reasonably assured by other means. However, relaxation of present siting criteria and/or containment philosophy require much more knowledge of the mechanisms and phenomenology of reactor accidents than now exists. Initial steps to provide such data were taken at ORNL in 1955. Early work was limited to the measurement of fission-product release from slightly irradiated metallic fuels. It was later extended to include ceramic fuels. Reports describing the work which was carried out prior to this reporting period are contained in the bibliography.

In July of 1961, at the request of the Division of Reactor Development, ORNL enlarged the scope of the Nuclear Safety Research and Development Program to include the assessment of safety problems of the entire reactor cycle. Current investigations are being made to provide information which will be useful in preventing - and assessing the consequences of - accidents which may occur in: reactors; fuel fabrication, shipping, and storage; fuel reprocessing; and fission-product separation and source production. The aim of the program is to obtain all of the data which are needed to evaluate the consequences of accidents, with emphasis on the study of those fission products which are limiting in the maximum credible accident for the part of the reactor cycle being investigated. At the present time, the ORNL Nuclear Safety Program encompasses the activities given below.

REACTIVITY EFFECTS DURING NUCLEAR EXCURSIONS

There is reason to believe that important geometrical changes may occur in plate-type fuel elements during a nuclear excursion. The resultant reactivity may be important in determining whether or not an accident ensues and, if so, its seriousness. The necessary data for resolving this question are being obtained by both experiment and computation.

REACTOR FUEL STORAGE AND HANDLING STUDIES

The safe fabrication, storage, and handling of reactor fuels require accurate data on the nuclear properties of the materials of interest. In the absence of accurate data, very conservative geometries, with resultant high costs, must be maintained during these operations. Therefore, data which will permit the specification of safe, realistic conditions during the fabrication, storage, and handling of fuel are being developed.

FISSION-PRODUCT RELEASE STUDIES

In order to evaluate the consequences of a reactor's maximum credible accident, detailed information is needed on the sequence of events following the accident. Both out-of-pile and in-pile fission-product release studies are being carried out to determine the identity and the fractional release of the fission products in the event of fuel meltdown or other catastrophe. The chemical and physical properties of the resulting aerosol are being established.

CHARACTERIZATION OF ACCIDENT-RELEASED FISSION PRODUCTS

In order to assess confidently the consequences of reactor accidents, a knowledge of the chemical and physical forms of materials which are released is necessary. The particles resulting from out-of-pile and in-pile meltdown and from oxidation of irradiated fuel samples are being characterized. The distribution of fission products between the gas phase and the particles is being determined, and the behavior of particulate matter in typical gas-cleanup operations is being assessed. Processes are being evaluated for removal of fission products from the containment-vessel atmosphere in order to lessen the hazard following a reactor accident.

NUCLEAR SAFETY PILOT PLANT

Before the fission-product release and transport data, developed on a laboratory scale, may be used with confidence, they must be validated under conditions closely approximating those which would prevail in the case of an actual reactor accident. The Nuclear Safety Pilot Plant, which is now in the design stage, will provide a facility in which the nonnuclear melting of irradiated fuel can be carried out on a larger scale than in the laboratory. The sequence of events - validation of fission-product

release, deposition, and reaction with structural materials; agglomeration and sedimentation of aerosols; and attenuation through leaks in the containment vessel - will be studied on a scale large enough to permit extrapolation to a reactor installation. Techniques will be evaluated for removing fission products from the containment-vessel atmosphere and for decontaminating the vessel itself.

REACTOR CONTAINMENT HANDBOOK

A complete collection of critically evaluated information needed for the design and testing of reactor containment structures is being prepared. In the Handbook, existing information will be evaluated and assembled into a suitable form for easy use by the reactor designer.

RADIOCHEMICAL PLANT SAFETY STUDIES

Radiochemical plants may suffer serious accidents of a magnitude comparable to those postulated for reactors. The nature of the materials which are released under conditions peculiar to radiochemical plants is being determined to provide a better understanding of the consequences of these accidents.

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PART I PHYSICS

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E. P. Blizard, Division Director
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2. REACTOR FUEL STORAGE AND HANDLING STUDIES

A. D. Callihan E. B. Johnson J. K. Fox

Experimental bases for the establishment of nuclear safety practices in the fabrication, storage, and transport of Rover Reactor fuel elements have been measured in a series of critical and neutron-multiplication experiments during this report period. This facet of the ORNL Nuclear Safety Program is nearing completion. Since the work is classified it will be described in a separate document.¹

¹E. B. Johnson and J. K. Fox, Critical Mass Studies, Part XII: Rover Reactor Fuel Elements, ORNL TM-264 (to be published).

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

E. G. Silver Z. M. Bartolome¹ K. M. Henry J. D. Kington

In order to be able to predict the magnitude of a power transient resulting from a rapid insertion of reactivity into a reactor, it is necessary to know the various mechanisms affecting the reactivity during a transient. Among these is the motion of fuel due to pressure effects. A program is currently in progress at the Bulk Shielding Facility to (1) determine the reactivity effects of small fuel displacements in the Bulk Shielding Reactor I (BSR-I) by experiment and (2) find adequate calculation methods whereby such effects can be computed. The experimental results are used to check the adequacy of the mathematical methods.

¹Affiliated with the Philippine Atomic Energy Commission; formerly on loan to ORNL.

EXPERIMENTS

A basic 25-element loading of the BSR-I reactor was perturbed by making compensating changes in the three center elements shown in Fig. 3.1. By "compensating change" is meant that as a fuel plate was removed two other specially made plates, each containing one-half the amount of fuel that was in the removed plate, were inserted in a water channel elsewhere in the three-element experimental region. Thus the total amount of fuel remained unaltered by the perturbation. In order to determine the effect of the proximity of a control-rod water-well, some loadings incorporated a nine-plate space in the center of the reactor core. In the majority of cases, the perturbations were designed to maintain the symmetry of the core as nearly as possible with respect to two mutually perpendicular planes (shown in Fig. 3.1) in order to facilitate comparison with the calculations. The indicated uncertainties in the results include possible errors in reading the regulating-rod position, uncertainties in the parameters of the inhour equation, and, most important, errors due

Fig. 3.1. Bulk Shielding Reactor I Core Configuration for Investigation of Fuel Displacement Reactivity Effects. (a) Top view of core configuration. (b) End view of special fuel element showing one plate removed and one $\frac{1}{2}$ -plate added. The representation of this element, as used in Fig. 3.2, is shown at the right.

to the free play in the fuel element positions and orientations. The last error was measured by several repetitions of measurements with intervening unloading and reloading of many fuel elements in order to introduce random changes.

The reactivity effects were measured by the use of a calibrated regulating rod (one of the four rods shown in Fig. 3.1) with a total worth of approximately 0.35% $\delta k/k$. This rod was calibrated by the use of the in-hour method, using the shim rods to compensate the reactivity of the rod being calibrated. In some cases the reactor configuration was subcritical with the 5×5 loading. In such cases two additional elements were added, as needed, in the positions indicated in Fig. 3.1. In this event the regulating rod was recalibrated to avoid errors due to changes in its importance. The results of all measurements (i.e., changes in reactivity) are shown in Fig. 3.2.

CALCULATIONS

Calculations were performed on some of the loadings tested by experiment, using several reactor codes which, in the past, had given satisfactory results for the critical mass of unperturbed reactors. To begin with, a one-dimensional calculation in cylindrical geometry was performed with the MODRIC Code,² a one-dimension, 32-group, diffusion-theory code which treats the hydrogen scattering in the Goertzel-Selengut approximation. This code yields energy fluxes which are then used to produce energy-weighted, few-group cross sections and diffusion coefficients. These few-group parameters (four groups) were used as input to the two-dimensional PDQ-2 Code,³ which is a straight, diffusion-theory, few-group code that solves the reactor equation in two cartesian or cylindrical dimensions. In order to improve the energy weighting in regions near discontinuities in the core, regions of homogeneous composition were subdivided into separate regions surrounding each interface; thus, the cross-section computation is separately carried out in the vicinity of such discontinuities.

In most cases, several MODRIC calculations were used to assemble the input parameters for the PDQ-2 calculation in order to take proper account of the effect of the eccentric location of some of the regions, which could not be adequately handled in a single, one-dimensional calculation.

The computed multiplication agreed with the experimental one within 1.5% in all cases, the calculated values lying systematically lower than the experimental results. Although this agreement is reasonably good, the interest here was in the relative differences between the perturbed and unperturbed reactor configurations. These differences, as calculated, are also shown in Fig. 3.2. It will be seen that, although the agreement seemed reasonably good in some cases, it was quite poor in others.

²MODRIC: A One-Dimension Neutron Diffusion Code for the IBM 7090, private communication of the Central Data Processing Group, Oak Ridge, unpublished.

³This is an IBM 7090 code which was adapted from a code used with the IBM 704 computer. The IBM 704 code is described in: G. G. Bilodeau et al., PDQ - An IBM 704 Code to Solve the Two-Dimensional Few-Group Neutron-Diffusion Equations, WAPD TM-70 (August 1957).

EXPERIMENT NUMBER	PLATE NUMBER																		$\delta k/k$ (%)	
																			MEASURED	CALCULATED
1	A																		(UNPERTURBED)	(UNPERTURBED)
	B																			
	C																			
2	A																		-0.092 ± 0.005	
	B																			
	C																			
3	A																		-0.207 ± 0.005	
	B																			
	C																			
4	A																		-0.314 ± 0.005	
	B																			
	C																			
5	A																		-0.422 ± 0.005	-0.301
	B																			
	C																			
6	A																		-0.061 ± 0.005	
	B																			
	C																			
7	A																		-0.140 ± 0.005	
	B																			
	C																			
7a	A																		-0.142 ± 0.005	
	B																			
	C																			
8	A																		-0.231 ± 0.005	
	B																			
	C																			
9	A																		-0.305 ± 0.005	-0.580
	B																			
	C																			
10	A																		UNPERTURBED WITH WATER SPACE	UNPERTURBED WITH WATER SPACE
	B																			
	C																			
11	A																		+0.025 ± 0.005	
	B																			
	C																			
12	A																		+0.081 ± 0.005	-0.158
	B																			
	C																			
13	A																		+0.056 ± 0.005	
	B																			
	C																			
14	A																		+0.016 ± 0.005	-0.163
	B																			
	C																			

CODE OF SYMBOLS

- | FULL FUEL PLATE IN NORMAL SLOT
- FUEL PLATE MISSING FROM SLOT
- λ 1/2 FUEL PLATE INSERTED BETWEEN NORMAL PLATES
- WATER SPACE

Fig. 3.2. Measured and Calculated Changes in Reactivity Due to Fuel Displacements in Various BSR-I Core Configurations.

Further calculations were performed to determine the cause of the disagreement. The results suggest strongly that the problem involves the transport effects that may be expected to be important close to region boundaries and in small regions of very different composition than that of the average core. The perturbations do constitute such regions. A one-dimensional, S_N -method calculation was performed for comparison with the MODRIC result. Figure 3.3 shows the two thermal flux shapes as obtained by the MODRIC and DSN codes (the latter is a one-dimensional, 18-energy-group transport code with four direction groups; it was used with the same mesh and input as the MODRIC case with which it is being compared). It is apparent that the effect of the water space and of the extra-fuel region is much larger in the S_N calculation.

Fig. 3.3. Comparison of Thermal-Neutron Fluxes Calculated by DSN and MODRIC Codes for a Cylindrical Model of a BSR-I Configuration.

Since there are, as yet, no transport-theory codes available in two dimensions, comparable to PDQ-2 in geometric sophistication, including transport effects, and coded for the IBM 7090, no direct testing of this hypothesis, as the source of the disagreement between the measured and calculated effects of these perturbations, has been possible.

CONCLUSIONS

The experiments indicate that negative reactivities result from all perturbations that decrease the homogeneity of the core, except when the perturbation adds fuel to high-importance regions, such as those adjacent to interior water volumes. However, there is not as yet enough evidence to establish these conclusions in general. Further experimental work, including larger regions of the core, is needed to accomplish this.

The calculational methods used appear to be inadequate to cope with the sort of small-dimension, large-magnitude perturbations represented by the experiments. No direct method of calculating these perturbations seems available in the absence of a two-dimensional transport code of sufficient scope to deal simultaneously with the multigroup, many-mesh point, multiangle problem, at least with acceptable computing times.

Further work is therefore planned in two areas, both involving experiments with special critical-experiment cores. For one, slab-type cores should be investigated with perturbations of the kind reported here in order to have a means to check the adequacy of the one-dimensional DSN code for geometries to which it is applicable. For another, there should be experiments with cores containing truly homogeneous-type perturbations, such as small changes in all of the water spaces in a given region of the core. Such experiments could be compared with diffusion-theory calculations, and if agreement is found in this case, then diffusion-theory codes could be used to predict the effects of perturbations of such a homogeneous type, which are indeed of practical interest.

PART II CHEMISTRY

REACTOR CHEMISTRY DIVISION

W. R. Grimes, Division Director

G. M. Watson, Coordinator

4. RELEASE OF FISSION PRODUCTS ON OUT-OF-PILE MELTING OF REACTOR FUELS

INFLUENCE OF IRRADIATION LEVEL ON FISSION PRODUCT HAZARDS ASSOCIATED WITH UO_2 -FUELED REACTORS

G. W. Parker G. E. Creek W. J. Martin R. A. Lorenz

Fission product release from irradiated UO_2 has been examined under out-of-pile conditions to permit maximum freedom in designing and conducting experiments. (Release studies performed under in-pile conditions are described in Chap. 5.) Studied so far have been diffusion of fission products into helium from solid materials at high temperature, release of fission products on oxidation of fuel material in air, release of fission products on melting of fuel in various atmospheres, and, finally, the effect of fission density or burnup on fission product release by each process. The effect of other variables, such as specimen size, environment, duration of heating, and overpressure, on the release of fission products during the melting of irradiated UO_2 will be considered in future studies.

Release of Fission Products on Oxidation of UO_2 to U_3O_8

A previous report¹ describes early results obtained in fission product release experiments performed with irradiated UO_2 . Some of the previously reported data are included in the present report for ready comparison with more recent data. The percent of fission product activities released from PWR-type UO_2 (93% of theoretical density), irradiated at tracer level, is plotted in Fig. 4.1 as a function of temperature. Note that a distinct discontinuity is observed at intermediate temperatures (800 to 900°C), which is also the temperature range in which the minimum oxidation rate for UO_2 in air occurs. This anomalous behavior is believed

¹G. W. Parker, G. E. Creek, and W. J. Martin, Chem. Div. Ann. Progr. Rept. June 20, 1961, ORNL-3176, pp 68-72.

to be associated with the occurrence of the maximum degree of plastic deformation of the U_3O_8 oxidation product at this temperature, thereby affording a maximum degree of surface-layer protection against further oxidation.

Release data obtained when UO_2 irradiated to 1000 Mwd/ton was heated in air to $1200^\circ C$ and when UO_2 irradiated to 4000 Mwd/ton was heated to a maximum temperature of $1200^\circ C$ are shown in Fig. 4.2 and in Tables 4.1 and 4.2 and for 7000 Mwd/ton in Table 4.3. Comparison of these release values with the data obtained with UO_2 irradiated at tracer level (Fig. 4.1) shows that, in general, release-rate values increase with increasing irradiation level. Data not reported here indicate that irradiated UO_2 oxidizes faster than unirradiated material under the same conditions. Since all the highly irradiated UO_2 material was received in the form of small fragments, these samples were made up of several small pieces having irregular shapes, while the tracer-level samples were single cylinders having smooth surfaces. The discontinuity observed at intermediate temperatures in Fig. 4.1 is missing

in most of the release curves shown in Fig. 4.2, but it is still observable in the curve showing behavior of iodine. It is probable that fission product impurity defects in the UO_2 and the greater surface area of irradiated samples led to a higher oxidation rate and thus to higher release values.

Fig. 4.1. Fission Product Release by the Oxidation of PWR-Type UO_2 to U_3O_8 in Air (Tracer-Level Irradiation).

Fig. 4.2. Fission Product Release by Oxidation of Highly Irradiated UO_2 (1000 and 4000 Mwd/ton).

Release of Fission Products from UO_2 by High-Temperature Diffusion

Rates of fission product release by diffusion from solid UO_2 at high temperatures should probably increase with increasing burnup in a manner similar to rates of release on oxidation of UO_2 . A series of diffusion experiments was conducted in which irradiated UO_2 specimens were heated in a stream of purified helium for 5.5 hr. An increase in irradiation

Table 4.1. Fission Product Release from UO_2 Oxidized in Air

Sample: Intermediate density PWR UO_2 (93-94%)

Irradiation: 1000 Mwd/ton

Air flow: 100 cc/min

Temperature (°C)	Time at Temperature (min)		Percentage of Individual Fission Products Released							
	He	Air	Rare Gases	I	Te	Cs	Ru	Sr	Ba	U
500		9	4.4	4.6	<0.014	0.02	0.013	<0.001	<0.0009	
	15	13	4.0	2.5	<0.003	<0.0008	<0.014	<0.004	<0.001	
	15	90	4.0	4.7	0.008	<0.002	0.36	<0.004	<0.0008	
600		11	6.6	3.4	0.003	0.004	0.33	0.003		
	15	13	6.0	5.6	<0.003	<0.0007	<0.23	<0.001	0.0009	
	12	90	5.5	6.0	0.005	0.003	0.9	<0.0009	<0.0009	
700		12	8.5	9.4	0.01	0.001	0.63	0.001		
	15	12	7.2	10.1	<0.003	<0.003	1.25	<0.0009	<0.001	
	13	90	8.3	10.0	<0.003	0.02	3.8	<0.0008	<0.0007	
800		15	10.5	9.1	0.05	0.016	5.2	0.0006		
	13	14	8.56	11.2	0.033	0.038	6.6	0.008	0.0006	
	13.5	90	15.1	14.1	0.08	<0.007	35.3	<0.001	<0.002	
900		15	11.2	15.2	<0.4	0.005	18.9	<0.001		
	10	14	11.9	14.4	<0.85	0.03	11.5	<0.002	<0.01	
	12.5	90	13.7	26.9	0.41	<0.002	30.3	<0.001	<0.0015	
1000		26	30.3	55.2	<7.7	0.07	81.5	<0.001	<0.007	
	10	18	22.2	42.2	<0.6	~0.03	69.8	<0.001	0.002	0.007
	12	90	30.5	73.3	31.3	0.02	97.9	0.002	0.005	<0.0012
1100	8	10	60.0	70.4	75.3	2.8	85.7	<0.001	<0.002	0.002
	12.5	11	49.9	64.1	28.0	<0.01	95.5	0.001	<0.016	
	12.5	90	52.7	71.2	58.0	<0.4	99.9	<0.2	<0.2	0.19
1200	17	15	79.6	86.6	59.4	<0.1	97.8	<0.03	0.1	
	14.5	90	77.0	83.4	75.9	4.5	99.7	0.14	0.14	0.142

Table 4.2. Fission Product Release from UO_2 Oxidized in AirSample: Intermediate density PWR UO_2 (93-94%)

Irradiation: 4000 Mwd/ton

Air flow: 100 cc/min

Temperature (°C)	Time at Temperature (min)		Percentage of Individual Fission Products Released							
	He	Air	Rare Gases	I	Te	Cs	Ru	Sr	Ba	U
500	16	23	1.5	3.6	<0.007	<0.0004	<0.005	<0.0004		
	18	90	2.9	3.2	<0.01	<0.0007	<0.01	<0.0004	<0.0008	
600	14	18	4.4	10.3	<0.006	0.002	0.075	<0.001		
	15	90	4.5	8.0	8.4	<0.001	1.8	<0.001	<0.004	
700	14	12	9.25	9.6	0.01	0.001	1.7	<0.0002	<0.0004	
	13.5	15	7.0	10.4	0.004	<0.001	0.44	<0.0003	<0.0006	
	14	90	6.8	6.5	<0.05	<0.0005	2.3	<0.0004	<0.002	
800	13	15	13.8	7.1	0.007	0.015	1.0	<0.0004	<0.0007	
	14	90	13.8	15.6	<0.06	<0.0096	11.9	<0.0004	<0.001	
900	14	19	20.5	48.9	0.4	0.009	17.3	<0.0001	0.01	
	15	90	21.5	47.1	6.0	0.015	53.1	<0.0008	<0.004	
1000	16	15	39.5	84.	11.5	0.09	72.1	<0.0003	<0.0016	
	13.5	90	43.8	75.	31.7	0.37	91.8	0.14	0.082	0.06
1100	14	14	66.0	79.4	15.9	<0.17	90.8	<0.05	<0.0032	
	14	90	73.4	83.6	38.6	0.19	99.3	0.006	0.014	<0.003
1200	14	16.5	71.3	81.7	37.1	0.76	89.9	<0.01	<0.001	
	13	90	79.7	94.5	66.4	6.4	99.6	0.0065	0.7	<0.0025

level was found to be accompanied by an increase in fission product release. Data from a PWR-type compact (93% of theoretical density) and an EGCR-type compact (97% of theoretical density), both irradiated to 1000 Mwd/ton, and similar data obtained at the tracer irradiation level and at the 4000-Mwd/ton level for PWR-type compacts are illustrated in Figs. 4.3 and 4.4. The data in Table 4.4 permit a more direct comparison of the actual release values at a given temperature. The burnup effect is shown to be quite marked for all fission products except perhaps ruthenium; the greatest effect is found in cesium release where differences of an order of magnitude are observed. Differences between the two types of fuel at the 1000-Mwd/ton level are not pronounced in spite of the density variation.

Table 4.3. Fission Product Release from UO_2 Oxidized in AirSample: High density PWR UO_2 (96%)

Irradiation: 7000 Mwd/ton

Air flow: 100 cc/min

Temperature (°C)	Time at Temperature (min)		Percentage of Individual Fission Products Released						
	He	Air	Rare Gases	I	Te	Cs	Ru	Sr	Ba
500	16	90	3.1	4.07	<0.5	0.0006	0.1	<0.0007	<0.0004
600	13.5	90	4.2	3.1	<0.013	<0.0017	0.73	<0.0004	<0.009
700	15.5	90	6.1	15.4	<0.079	<0.0046	0.112	<0.0005	<0.007
800	14	90	9.4	9.0	<0.3	0.002	9.8	<0.0005	0.03
850	13.5	90	14.6						
900	16.5	90	33.9	29.3	80.4	<0.012	78.0	<0.03	<0.8
1000	16.5	90	86.4						
1100	Incomplete								
1200	Incomplete								

Table 4.4. Effect of Burnup Level and Density on Diffusion of Fission Products from Irradiated UO_2 into Pure Helium at Various Temperatures

Temperature (°C)	Irradiation Level (Mwd/ton)	Percentage of Individual Fission Products Released ^a						
		Xe-Kr	I	Te	Cs	Ru	Sr	Ba
1400	Tracer	0.8	4.0	3.9	0.02	0.02	0.001	
	1005 (EGCR)	0.8	0.9	0.8	2.6	0.001	0.1	
	1000 (PWR)	0.5	1.6	1.2	0.5	0.001	0.06	1.8
	4000 (PWR)	6.1	23	16	21	0.006	0.08	0.5
1610	Tracer	2.7	6.5	12.1	1.7	1.5	0.1	
	1005 (EGCR)	2.6	3.7	12.0	12.0	0.1	2.0	17.0
	1000 (PWR)	6.0	5.5	27	20	0.3	0.2	12
	4000 (PWR)	14	25	48	43	0.2	0.5	15
1780	Tracer	3.7	11.7	21.0	3.2	6.9	1.0	
	1005 (EGCR)	12.0	24.0	67.0	27.0	0.4	9.0	39.0
	1000 (PWR)	14	25.5	35	22	0.4	3.7	21
	4000 (PWR)	42.2	59.4	59.8	39.7	5.7	5.8	18.4
1980	Tracer	12.3	41.0	75.0	15.0	13.4	4.2	8.7
	1005 (EGCR)	29.0	53.0	74.0	84.0	6.0	15.0	57.0
	1000 (PWR)	48.5	63	90	69.5	4.8	~10	51
	4000 (PWR)	71.3	80.8	81	98.3	14.9	33.0	60.4

^aIncludes that portion adsorbed on crucible and reflector parts.

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Fig. 4.3. Release of Fission Products by Diffusion from Highly Irradiated PWR-Type UO_2 Heated 5.5 hr in Purified Flowing Helium.

Fig. 4.4. Fission Product Release from Highly Irradiated EGCR UO_2 and from Trace-Irradiated PWR UO_2 by Diffusion into Pure Helium.

Release of Fission Products from UO_2 Melted in Helium, Air, and CO_2

The usual hazards analysis for a loss-of-coolant accident to any large water-cooled reactor postulates melting of much of the fuel loading by decay heat. These hazards analyses unfortunately do not usually take into consideration the duration of the molten period, the velocity of movement of the gas cloud formed in the accident, or the composition of the gaseous atmosphere present during melting. It seems logical to assume that a large mass of UO_2 probably would not remain molten long. The material would probably flow out of the region of highest temperature under the influence of gravity, and it would soon solidify and entrap the remaining fission products. The melting experiments have accordingly been designed to maintain UO_2 in the molten condition for short periods in various atmospheres such as helium, air, and CO_2 . The most important condition for which no data have yet been obtained is a simulated meltdown in a steam atmosphere; however, it is felt that the release results obtained in steam would not differ greatly from those obtained in helium or CO_2 .

The data in Table 4.5 show the effect of atmosphere and burnup on release of fission products during the melting of UO_2 . Since the release values for the volatile fission products are ordinarily quite high even on melting tracer-level-irradiated UO_2 , the burnup effect is not especially noticeable in these data.

Table 4.5. Effect of Burnup Level and Atmosphere on the Release of Fission Products During the Melting of UO_2 ^a

Atmosphere	Irradiation Level (Mwd/ton)	Sample Weight (g)	Number of Results	UO_2 Vaporized (%)	Percent of Individual Fission Products Released							
					Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Rare Earths
Helium (impure)	Tracer	0.22	2		99.5	89.7	92.0	91.3	61.0	2.1	4.6	2.2
	2,800	0.03	3	21.2	99.9	92.2	98.2	98.5	90.4	2.1	6.6	5.1
	11,000	0.03	2	18.0	100.0	99.7	99.6	98.7	88.1	1.7	6.5	4.1
Air	Tracer	0.2	2		98.4	94.9	79.1	37.7	67.7	0.2	0.5	0.5
	2,800	0.04	3		100.0	99.7	93.6	92.5	95.0	0.4	1.8	3.0
	11,000	0.04	3	24.7	100	99.8	99.3	97.4	92.5	0.4	1.8	3.9
CO_2	Tracer	0.2	3	14.1	80.6	76.8	71.2	60.9	44.9	0.3	1.1	0.85
	2,800	0.02	3		99.9	98.7	98.6	90.2	74.3	0.5	2.5	2.8
	11,000	0.05	3		99.9	99.9	99.0	96.6	79.1	0.6	2.9	2.3

^aEGCR UO_2 with an O/U ratio of 2.04 and an average density of 95% of theoretical.

Production and Deposition of Particles on Melting of UO_2

The size of particles released in a reactor accident affects the fraction of the released activity which will be deposited within the reactor containment shell before it reaches holes in the shell and the fraction of the remaining activity which will be deposited during passage through the opening. In Fig. 4.5 a particle-size analysis is given for the released uranium oxide particles volatilized in different atmospheres. Most of the particles had diameters ranging from 0.01 to 0.1 μ . An electron-microscope photograph of some of these particles is found in a previous report.¹

An indication of the range in quantity of material which may be plated out (before reaching an absolute filter) as a function of the velocity of air passed over the sample during melting is given by the data in Table 4.6. Of particular interest is the range in quantity of iodine found to pass through the apparatus and filters and to be collected finally in activated charcoal. The data demonstrate that while most of the refractory fission product oxides are distributed to some extent in proportion to the amount of uranium oxides deposited, the more volatile iodine may be carried considerable distances by a rapid flow of gas. Up to 10% of the uranium may volatilize in a typical melting experiment, while an average of less than 1% of the strontium is volatilized in the process. This fact is very encouraging from the reactor hazards viewpoint.

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Fig. 4.5. Particle-Size-Evaluation Oxides Vaporized from Melted UO₂ (Arc-Image Furnace).

Table 4.6. Effect of Furnace Air Velocity on Deposition of Fission Products Released by Melting Modified UO₂ Specimens at 2650°C

	Percent of Total Activity and Its Location, with Velocity of Furnace Air as Indicated (lin ft/min) ^a							
	Furnace Tubes		Filters		Charcoal Trap		Total	
	0.3	5.0	0.3	5.0	0.3	5.0	0.3	5.0
Gross gamma	10.1	8.8	1.2	6.2	0.4	3.4	11.7	18.4
Rare gases					77.1	69.5	77.1	69.5
Iodine, γ	62.3	12.1	5.4	9.8	11.3	56.9	79.0	78.8
Tellurium, β^-	66.8	28.6	8.3	43.2			75.1	71.8
Cesium, γ	34.5	18.3	15.6	30.9			50.1	49.2
Ruthenium, γ	63.6	33.0	0.8	24.6			64.4	57.6
Cerium, β^-	0.24	0.17	0.11	0.37			0.35	0.54
Strontium, β^-	0.1	0.05	0.01				0.11	0.05
Barium, β^-	0.55	0.08	0.01	0.07			0.56	0.15
Zirconium, γ	0.02	0.02	0.01	0.01			0.03	0.03
Uranium ^b	6.4		1.4				7.8	

^aAir flow = 200 cc/min or 0.3 lin ft/min or 3000 cc/min or 5 lin ft/min across sample; heating time = 90 sec.

^bVaporized as UO₃.

UO₂ MELTDOWN DEVELOPMENT PROGRAM

A development activity has been initiated as a part of the out-of-pile fission-product-release program, in order to afford a more realistic simulation of the loss-of-coolant meltdown. A critical analysis of the difficulties encountered in applying a large scale-up factor to fission-product-release data based on small laboratory-scale melting of UO₂ leads to the conclusion that little satisfaction can be gained from the present procedure because of the great differences between the conditions existing in the laboratory experiments and in an actual reactor accident. While developments have already been reported for melting capsules containing up to 80 g of irradiated UO₂, these developments are being delayed by difficulties in the irradiation program, and the proposed experimental system is inherently subject to significant limitations (as are most meltdown experiments) related to the containment of the melt in a crucible. Geometrical confinement of the melting fuel undoubtedly reduces the escape of fission products according to some function of the solid angle for dispersal and of the chemical reactivity of the crucible. In the arc-image-furnace melting process, the solid angle for dispersal is obviously at a maximum. In a reactor meltdown accident, following the loss of cladding, the solid angle for dispersal on melting would vary depending on the mode of melting and the degree of mechanical disruption of fuel geometry by fragmentation and other possible means. These factors can only be appraised in larger experiments. This reasoning has led to a reevaluation of experimental UO₂ meltdown and to a search for new containment materials. A major advance in melting technique resulted from the discovery of the compatibility of molten UO₂ with metallic tungsten. This technique furnishes a means of melting UO₂, not only in large quantity, but also under the more realistic condition of center melting of a fuel pin. In order to achieve the latter condition, the UO₂ is cored and heated to the melting point from the center outward by means of a tungsten-rod resistance element supplied with suitable low-voltage high-amperage power. Unclad UO₂ can also be melted in an inert atmosphere by coupling inductively to a tungsten crucible. These methods are described below.

Tungsten Crucible Method for Induction Melting of UO₂

A number of advantages were found to be associated with the use of tungsten-foil crucibles which were readily heated by rf induction to the melting temperature of UO₂. Particular interest is attached to the fact that a scale-up factor of 1000 was readily attained over the maximum size in the arc-image-furnace method which was previously used for the melting of UO₂ and had been found to be limited to samples of less than 50 mg of highly irradiated UO₂. Tungsten crucibles as shown in Fig. 4.6 made from 5-mil tungsten foil may be used to melt 50 g of UO₂ of any burnup level obtainable. This factor of 1000 is expected to result in a significant reduction of the nearly quantitative releases indicated for the more volatile fission products by the arc-image-furnace melting data.

The use of tungsten at high temperatures is limited to noncorrosive atmospheres. However, the previous fission-product-release data for melting UO₂ indicates little or no atmosphere effect; therefore, it is probably not unrealistic to conduct release experiments in an inert gas such

as helium. Some corrections may be desired in order to extrapolate to melting in steam or in steam and air mixtures, but these correction factors are not expected to be large. The melting behavior of UO_2 in the tungsten crucible is illustrated in Fig. 4.7 in which the concave meniscus of the surface of solidified UO_2 is seen from the top of the crucible. Bright, smooth, reflective under-surfaces are seen on a few incompletely melted pieces which remained above the surface of the fluid UO_2 . The upper sides of these fragments show considerable evidence of crystal growth by deposition from the vapor phase as does one side wall of the crucible.

Fig. 4.6. Tungsten Crucibles Showing Varied Appearance of Induction-Melted UO_2 . At left a melt with smooth top surface heated slightly above melting, and at right a sample with crystal growth on the surface heated slightly below melting. Thermowells for tungsten-rhenium couples are shown.

Fig. 4.7. Sample of Melted UO_2 Showing Smooth Bright Undersurface of Residual Fragments Remaining on Side Walls and Melted on Bottom Only. Tops show crystal growth by evaporation.

Upon melting, a prompt release of UO_2 vapor is observed, which appears as a burst of relatively short duration. The resulting smoke when conducted through a small chamber, as shown in Fig. 4.8, leaves a deposit of the major portion of the UO_2 in the front section of the chamber. The gas flow rate, which is determined by the quantity of helium entering the system, provides a relatively low velocity of 1 fpm across the chamber. Vaporization of UO_2 during a 5 min melting period resulted in a loss of about 1% of the total uranium, about 90% of which plated out in the hot zone. An absolute filter at the end of the chamber collected from 1 to 2 mg of UO_2 particles from a 35 g melt. This amounts to less than 0.01% of the UO_2 sample.

Fig. 4.8. Induction-Furnace Tube and Miniature Fallout Chamber Used with Tungsten-Crucible Method for Melting UO_2 . Heavy plate-out of UO_2 in the furnace hot zone is evident.

Tungsten-Rhenium Thermocouple Measurements of UO_2 Melting Temperatures

In some of the experiments in which UO_2 was melted in a tungsten crucible, an inverted thermowell was built into the crucible permitting introduction of a tungsten vs tungsten-26% rhenium thermocouple. The observed melting temperature of UO_2 was 2830°C . It is believed that the melting temperature can also be observed readily for UO_2 with high burnup, in order to verify the expected melting point lowering of UO_2 by fission products. A calibration experiment for the thermocouple resulted in the curve shown in Fig. 4.9.

Fig. 4.9. Calibration Curve for Tungsten-Rhenium Alloy Thermocouple and Observed Melting Point for UO_2 Heated in a Tungsten Crucible.

Meltdown of UO_2 Fuel Pins by a Center Tungsten Resistor

Center melting has been one of the most difficult fuel conditions to simulate in a nonnuclear heating experiment. A simple mockup of these conditions was produced by the construction of the resistance heated assembly shown in Fig. 4.10. A metal-clad capsule containing cored PWR UO_2 fuel pellets, through which a tungsten rod is passed, is surrounded by a glass container which can be evacuated and filled with an inert gas. Electric heating is accomplished by passing a low-voltage current through the tungsten rod from water cooled terminals. With a helium atmosphere and an aluminum oxide reflector tube, a three-pellet assembly melted, as shown in Fig. 4.11. Since the outer UO_2 surface is radiating to relatively cold surroundings, the skin of the fuel pellet as observed does not melt freely. Examination of a cross section of the specimen, however, showed the unmelted UO_2 to be a very small fraction of the sample. Melting of a five-pellet assembly is shown in Fig. 4.12. The horizontally extended spurs of solidified UO_2 were found to be hollow suggesting that they were forced out by internal gas pressure. This also explains the upward extrusions at the top of the assembly. Identification of this gas, which is probably oxygen, is being planned.

Projected Scale-Up of the Center-Melting Technique to a Cluster Meltdown

The most-nearly-realistic, accident-oriented, fission-product-release experiment will involve an assembly of fuel pins into a cluster so that the heat radiating from the outer pins is conserved to effect complete melting of the inner pins. Such an experiment is being designed in its simplest form as a five- or seven-pin cluster. In this case, complete

Fig. 4.10. Vacuum Chamber and Electrode Assembly of Tungsten Resistor for Melting Cored- UO_2 Fuel Pin Capsule.

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Fig. 4.11. Sample of Melted PWR UO_2 in a Three-Pellet Capsule Showing Melted Cladding Below and Extruded UO_2 on Sides and Top.

melting would occur only in the center pin. A demonstration of fission-product release from an assembly of this type will include in one experiment the simulation of all the more important effects of size and physical arrangement. This will permit an extrapolation to a large-scale release with confidence.

Power-input requirements are expected to be about 20 kw or 20 w per g of UO_2 . While a rare gas is the only practical atmosphere for use at the present time, there is a possibility that steam may be used in later experiments if it is found to be compatible with this system.

Fig. 4.12. Sample of a Five-Pellet Capsule Melted by the Center Tungsten Resistor Showing Most of Melt Expelled at the Bottom. Evidence of interior gas pockets is seen near horizontal and upward UO_2 extrusions.

Scale-Up of Parameter Studies to 80 g of UO_2
in Metal-Clad Fuel Pins

One long-standing program in which progress is being made is concerned with the melting of zirconium-clad (Dresden) and stainless-steel-clad (SAVANNAH) UO_2 fuel in pins of various lengths in order to evaluate the following effects:

1. Metal cladding (stainless steel or Zircaloy-2),
2. burnup (0-10,000 Mwd/ton),
3. atmosphere (steam, air, helium).

Since all the actual high-level work must be accomplished in hot cells, an extensive design program has been necessary in order to prepare for this scale-up in quantity of radioactivity.

Induction heating has been the method of choice for the fuel since it lends itself to melting first the cladding and then the UO_2 . A detailed irradiation program has been necessary to provide the necessary burnup in the 60 capsules in the program. Figure 4.13 shows the general

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Fig. 4.13. Layout of Hot Cell and Fallout Chamber for Induction Melting UO_2 Capsules.

layout of the hot cell for the melting. Additional features of the cell include a large glass chimney above the furnace tube to conduct the smoke and fumes into the plate-out chamber.

In this apparatus some analyses of particle behavior as well as iodine "wash-out" will be conducted. Additional details of this apparatus are given in Chap. 6.

A second hot cell has been assigned for handling the chemical recovery and sampling for analytical material balance operations in order to obtain release values from the melting experiments conducted in the adjacent hot cell.

Preliminary experiments have shown that a minor difference already exists in the behavior of the two metal claddings. For example, in Fig. 4.14, stainless steel cladding is shown to produce a segregated metal button which tends to flow further away from the solidified UO_2 . Zircaloy-clad UO_2 apparently forms a single phase of $\text{UO}_2\text{-Zr-ZrO}_2$ as is shown in the

Fig. 4.14. Samples of Induction Melts of UO_2 (Left, Stainless Steel Clad, and Right, Zircaloy Clad) Showing Differences in Segregated Stainless Metal and Single-Phase $\text{Zr-ZrO}_2\text{-UO}_2$.

figure for comparison. Since ZrO_2 is not a normal reactor material, some other material may be used in subsequent experiments in order to avoid this complication.

Design for Melting UO_2 in TREAT

Since some parameters worthy of investigation (such as overpressure) either cannot be accomplished with ease under other conditions or are only associated with a fast reactor transient, some attention has been directed toward conducting release experiments by melting clad fuel pins in the Transient Reactor Test (TREAT).

A great number of UO_2 meltdown experiments have already been carried out in this facility by Argonne personnel as a part of the metal-water reaction program. The results of these experiments seem to indicate a need for heating the UO_2 specimens to an annealing temperature prior to the application of nuclear heat, in order to eliminate fragmentation of the fuel specimen during the reactor transient. Consequently, preliminary heat balances have been conducted to evaluate the insulating qualities of

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Fig. 4.15. Double Autoclave Assembly Concept for TREAT In-Pile Melting.

concentric tungsten-foil heat shields for insulating the fuel capsule in the present concept of a twin autoclave arrangement which is shown in Fig. 4.15.

The purpose of the twin autoclave is to sample the smoke released following a transient meltdown. The cavity in the second vessel is evacuated and sealed with an explosive valve which opens downstream to a set of absolute filters. A controlled-rate transfer of gas to this cavity will collect half the volume of gas contained in the first autoclave.

Highly irradiated fuel will be melted in the TREAT facility after preliminary melting experiments with unirradiated fuel specimens are completed. Hot-cell dismantling and examination is being provided.

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

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A series of in-pile experiments is being conducted in which miniature fuel elements are melted by fission and gamma heat. These experiments are being performed to study the fission product release during a simulated reactor accident in which the fuel elements are destroyed by melting or vaporization as a result of uncontrolled heat generation or loss of reactor coolant. Since the fuel specimens are heated internally as a result of neutron and gamma fluxes, these experiments simulate reactor accident conditions, in this respect, more realistically than do out-of-pile experiments.

In a typical in-pile meltdown experiment, a fuel specimen is placed in a reactor furnace (described below) and moved to a position close to the top of the lattice of the Oak Ridge Research Reactor (ORR) to build up traces of fission products. After an irradiation period the specimen is lowered to a position in the lattice where fission and gamma heat causes it to melt. Helium gas sweeping over the specimen carries gas-borne fission products to suitable filters and traps, where they are separated and collected. After the specimen is withdrawn from the flux, the furnace, filters, traps, and associated tubing are removed from the reactor. These parts are then cut up in a hot cell and examined, and finally are analyzed for fission products.

The details of the reactor furnace are shown in Fig. 5.1. Specimens in the initial series of experiments were stainless-steel-clad UO_2 cylinders, 0.210 in. in diam. by 1 in. long, and had a density of about 95% of theoretical. The specimen in each experiment was mounted on a porous thoria holder which was supported by a porous zirconia base resting on a beryllia heat conductor. The specimen region was enclosed by a thoria

Fig. 5.1. Reactor Furnace for Meltdown Experiments.

cylinder and had a thorium roof plug and was surrounded by a zirconia cylinder. The thorium served as a container for the molten UO₂, and both the thorium and zirconia served as heat insulators. These parts were all contained in a primary stainless steel container which was enclosed in a secondary stainless steel container. Both the top and bottom of the primary container had aluminum-filled caps to transfer heat from the ends of the primary container to the side of the secondary container. The experiment was cooled by reactor tank water flowing past the secondary container. The integrity of the system was monitored by stagnant helium pressurized above reactor tank pressure. Leaks in either containment system were detected by a drop in the helium pressure.

During the time an experiment is in the reactor, a flow of helium (see Fig. 5.2) passes through the primary system and carries particulate matter and fission products from the furnace through a stainless steel

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Fig. 5.2. Experimental Flow Diagram.

tube to three filters in series and a charcoal trap. The filters retain particulate matter, and the charcoal, maintained at reactor tank water temperature (120°F), removes iodine and possibly other fission products from the gas stream. The sweep gas then passes through a liquid-nitrogen-cooled charcoal trap located outside the reactor pool. This charcoal retains all the rare gases released from the irradiated fuel specimen. Radioactive gases in the inlet and exit lines to this trap are monitored by ion chambers connected to recording electrometers in the experiment control room. From the cooled-charcoal trap the helium sweep gas flows into the reactor off-gas system.

Experiments are installed in a permanently mounted facility tube (Fig. 5.3), which enters the ORR top plate through the V-5 access flange and is bent to pass into the F-9 lattice position. The lower end of the facility tube fits into a lattice insert which is machined to fuel element dimensions. A hydraulic positioning mechanism is mounted on the top of the facility tube for the purpose of lowering the furnace into the reactor lattice or raising it to a position above the lattice while the reactor is operating. This mechanism can move the experiment up or down over a 36-in. range. Solenoid valves operated from the control panel direct the flow of process water which operates the mechanism.

A pipe which penetrates the top of the reactor tank through the flange supporting the facility tube permits tank water to be directed by

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Fig. 5.3. Fuel Meltdown Experiment Assembly in Reactor.

a pair of mechanically linked valves either into the facility tube to provide a cooling medium for the experiment or alternatively through a pipe which runs to the bottom of the facility tube to operate a hydraulic plunger to close a foot valve in the cooling water outlet. When the mechanically linked valves are in the latter position, the facility tube is isolated from the reactor tank, and the tube may be opened either to insert or to remove an experiment while the reactor is operating.

Five experiments have been performed. The first three of these experiments were designed to test the operational characteristics of the design. Experiment 1 contained 3% enriched fuel and produced a maximum power of approximately 60 w/g. The second experiment contained 25% enriched fuel and produced a power of approximately 120 w/g at the point where the UO_2 melted. Experiment 3 contained 6.5% enriched fuel and produced a maximum power of 120 w/g at the fully inserted position. During these three experiments the equipment functioned as designed. Installation and removal of the experiments were made during reactor operation. Heat transfer calculations were verified showing that the necessary temperature could be reached inside the furnace while keeping the container cool. Radiation levels at the trap collection points were within acceptable limits. And most importantly, it was shown that controlled melting of UO_2 could be accomplished in-pile. Under the prevailing conditions, temperatures estimated to be about 3500°C have been produced.

Experiments 4 and 5 were the first two of a series of experiments designed to test the reproducibility of the experiments. Each contained the same fuel as expt 3 and conditions, such as insertion time and duration of melting, were maintained constant.

Examination of material from expt 1 in hot cells showed the UO_2 specimen to be intact (Fig. 5.4) but to contain a large central void over $3/4$ of its length. The outer surface of the specimen consisted of pyramidal crystals, shown in more detail in Fig. 5.5. Figure 5.6 is a photomicrograph of a section of the UO_2 . The dashed line shows the estimated position of the original circumference of the specimen. There are large

Fig. 5.4. Segment of UO_2 Specimen from Expt 1 After Being Heated Internally to the Melting Point.

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Fig. 5.5. Crystalline Outer Surface of UO_2 Specimen from Expt 1.

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Fig. 5.6. Photomicrograph of Section of UO_2 Specimen from Expt 1. The dashed line indicates the original surface of the specimen.

grains and voids visible, but the columnar growth often observed in UO_2 which has been subjected to high-temperature gradients is missing. The sharp well-defined crystals on the surface indicate that a considerable quantity of the UO_2 has passed through the pores and cracks to the outside as vapor, where it crystallized on the cooler surface. The temperatures required to produce the effects displayed in these figures are consistent with the temperatures found by extrapolation from the temperature observed during irradiation.

In expt 2 the fuel specimen had vaporized or melted completely, and crystalline UO_2 had collected throughout the inside of the ThO_2 cylinder, as shown in Fig. 5.7. In Fig. 5.8 penetration of the UO_2 into the ThO_2 holder, which was evident upon metallographic examination, is shown. The unirradiated ThO_2 holder is shown for comparison.

In expts 3-5 the residues of the UO_2 specimens were solid masses apparently resulting from solidification of molten puddles. No crystal formation was evident. As an example, the ThO_2 holder from expt 3 is shown in Fig. 5.9.

Analysis of the material inside the primary containment of expts 2-5 shows that nearly all the UO_2 remains inside the furnace. However, in each experiment, between a third and a half of the UO_2 has been transferred from the holder region to other regions of the furnace.

Radiochemical analysis for various fission products has been performed and the data are now being calculated and interpreted. Although only a small fraction of the data are available at this time, some interesting trends can be seen. Table 5.1 gives a measure of the homogeneity of the fission product distribution in the UO_2 . Each number is the ratio of the amount of the fission product found in the UO_2 transported to the

Fig. 5.7. ThO_2 Holder from Expt 2 Showing Hole Where ThO_2 Melted or Dissolved in UO_2 .

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Fig. 5.8. Comparison of Unirradiated Holder with Holder from Expt 2 Showing Penetration of UO_2 into the Holder.

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Fig. 5.9. ThO_2 Holder from Expt 3 Showing Solidified Melt of UO_2 .

Table 5.1. Homogeneity of Fission Products^a in UO_2

Isotope	Distribution Ratio	
	$\left(\frac{\text{FP in } \text{UO}_2 \text{ on walls}}{\text{FP in } \text{UO}_2 \text{ in holder}} \right)$	
Ru^{106}	0.992	
Ce^{144}	0.867	
Sr^{89}	0.745	
Zr^{95}	0.647	
Ba^{140}	0.516	
I^{131}	0.337	

^aOrder of increasing vapor pressure at 2200°C: Ru , ZrO_2 , BaO , SrO , Ce , Ba , Sr .

cylinder walls to the amount found in the UO_2 remaining in the holder. For comparison, the order of increasing vapor pressure of the elements is also shown. Apparently, as the UO_2 travels, it tends to lose iodine but retains ruthenium.

Some further observations, which can be made concerning these data, are that Ce^{144} is retained to a greater extent than Ba^{140} and that Ba^{140} and Sr^{89} behave similarly. Iodine and the rare gases krypton and xenon are released to the greatest extent. The rare gases are used as fission monitors. The data also show that the greatest amount of total release was accomplished in expt 2, which was irradiated without cladding, in which crystalline deposition of fuel occurred, and which was held molten for the longest time.

Interpretation of the analytical data for expt 3 is nearly complete. Satisfactory material balances were obtained for individual fission-product isotopes. Table 5.2 lists the total amount of each isotope found, expressed in terms of the equivalent number of fissions and the fraction of each found inside the high temperature furnace. It is significant that although a large fraction of the fission products escape from UO_2 during melting, they are almost entirely retained, except for the rare gases, inside the high-temperature zone of the experiment.

Experiments to be performed in the immediate future will continue the reproducibility series. These will be followed by experiments in which conditions, such as duration of melting, atmospheric environment, gas velocity, and fuel material, are varied.

Table 5.2. Fission Products Obtained in Experiment 3

Isotope	Number of Fissions Equivalent to Amount Found	Percent Retained in Furnace
	$\times 10^{17}$	
Sr^{89}	3.24	98.9
Zr^{95}	3.36	99.9
Ru^{106}	3.59	99.9
Ba^{140}	5.15	99.5
Ce^{144}	3.50	97.0
Rare gases	3.48	0

6. FISSION PRODUCT TRANSPORT EVALUATIONS

C. J. Barton

G. W. Parker

The purpose of this study is to provide detailed information concerning the changes that occur in the size distribution of particles released in simulated fuel meltdown accidents and in the chemical form of the fission products carried by these particles while they are being transported by gases. Such information is needed in order to obtain a better understanding of the many factors which would affect the sedimentation of released fission products within reactor containment vessels and the fraction of these hazardous materials that may escape to the environment. Since the present program was started recently, little factual information is now available, but this progress report will indicate the contemplated approach to the problem.

PARTICLE SIZE STUDIES

Some data have been obtained on particle size distribution of particles released in out-of-pile melting experiments (see Chap. 4). The small size of the laboratory equipment in which these experiments are customarily performed is not conducive to the extrapolation of the results obtained to the conditions existing in actual power-reactor accidents where released fission-product particles in the containment shell would have an appreciable amount of time to plate out, react with gases in the atmosphere, or undergo other physical or chemical changes. To help bridge the gap between present laboratory experiments and full-size reactors, a diffusion, or fallout chamber, is being constructed for use in conjunction with scaled-up melting experiments. Initial tests with cold or tracer-level materials will be performed with the box shown in Fig. 6.1, constructed with an aluminum frame and Lucite sides. These experiments will be carried out in a hot cell, because of convenience and safety considerations, with a dual-frequency generator to heat clad UO_2 specimens in oxidizing atmospheres. Several feet of glass tubing, a part of which will be in the vertical position, will be required to conduct the gas stream from the heated zone to the fallout chamber, and, consequently, large particles are not expected to reach this part of the apparatus except with very high gas flow rates. However, since particles having a diameter of 1.0μ or less are most likely to remain gas-borne long enough to escape from a containment shell, the behavior of these small particles will receive major attention in the present program.

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Fig. 6.1. Diffusion Chamber for Study of Particles from Melted or Oxidized Irradiated Fuel.

Several techniques will be employed in the study of the small particles that deposit during passage through the fallout chamber, in addition to the gas-borne particles passing through the chamber. Some particles will be collected on Millipore filter paper or a similar material, in order to facilitate examination by the electron photomicrographic technique. Such samples will also be scanned with a multichannel analyzer for qualitative identification of radioactive species present. Quantitative data on relative amounts of these species will be obtained by radiochemical analysis of materials deposited in trays in various parts of the chamber. A number of techniques are described in the literature for determining the size distribution of gas-borne particles. Recent data on the deposition of T^{131} -tagged particles in constricted-channel diffusion cells, reported in Chap. 7, makes use of this technique attractive for the study of particles produced when UO_2 is melted in different atmospheres. It is expected that one or more of similar diffusion cells will be employed in the present investigation.

A stainless steel fallout chamber has been designed for the study of particles released on melting comparatively large quantities (up to 80 g) of highly irradiated UO_2 in a hot cell. This design is subject to modification on the basis of information obtained with the aluminum-Lucite chamber, shown in Fig. 6.1, and it will be described in a subsequent report.

IDENTIFICATION OF CHEMICAL SPECIES

Although some progress has been reported in the identification of the chemical form of a few released fission products, no technique has been reported which is capable of unambiguous identification of a variety of chemical species. One purpose of this program is to develop such a technique.

One possible means of achieving this aim is to make use of the fact that different chemical compounds, in which an element may exist, usually have different vapor pressures. The apparatus shown in Fig. 6.2 was constructed to test this possibility with a radioactive species which is of general interest to reactor safety, and which exists in both volatile (molecular I_2) and nonvolatile forms (various iodides and iodates). A small amount of molecular nonradioactive I_2^{127} (1 or 2 mg) tagged with about 0.1 mc of I^{131} is sealed in a small glass ampoule and placed in the support funnel which is then inserted in the tube which is surrounded by heating tape. A small charcoal trap is placed in the upper part of the glass tube, a slight vacuum (indicated by a mercury manometer) is applied at the top end of the tube, and a measured gas flow is introduced at the lower

Fig. 6.2. Iodine Transport Apparatus.

end of the tube. After checking the background count with the sealed ampoule placed in the apparatus, the thin top window of the ampoule is broken by manipulating the stainless steel rod inserted in the apparatus through a flexible brass bellows. The activity on the small charcoal trap is measured at regular intervals by means of a crystal detector connected to a decade scaler and is simultaneously measured and recorded by a linear-rate count meter. The temperature inside the tube is measured by means of a thermocouple connected to a portable potentiometer.

Two exploratory experiments completed to date have served to demonstrate that the experimental arrangement should give useful information on the behavior of iodine as a function of temperature, gas flow rate, and gas composition. Figure 6.3, which is a plot of I^{131} activity on the charcoal trap at various times in the second experiment, shows that a considerable amount of iodine is carried by helium at room temperature followed by a slower, more or less linear, rate of transfer. Increasing the temperature increased the transfer rate, as might be expected, and, at three temperatures, there was a short period in which a constant rate of increase in I^{131} count was observed, indicating that the I^{131} concentration in the sweep gas was constant during this period, since the gas flow rate was constant. An approximately straight line was obtained by plotting the logarithm of the rate of increase in the I^{131} count, which is the slope of the I^{131} count curve, vs the reciprocal of the absolute temperature. A similar linear relationship in the data plotted in this manner

Fig. 6.3. Iodine-131 Transported by Various Gases and Collected on Charcoal.

was observed when moist laboratory air was pulled through the apparatus at various temperatures, following removal of a large fraction of the iodine activity by passage of helium and helium I_2^{127} through the apparatus in the first experiment. The order in which the data points were determined apparently did not affect the values obtained, but one point obtained using compressed air was significantly lower than the line representing the data obtained with laboratory air, possibly due to the lower water content of the compressed air. These tests also confirmed earlier reports,¹⁻³ that I^{131} adsorbed on a surface can be removed by exchange with inactive iodine, but further tests under more carefully controlled conditions will be required to determine the extent and rate of exchange of active and inactive iodine. Since surface reactions undoubtedly play an important part in the desorption of iodine from surfaces, it will be necessary to study this phenomenon in the absence of carrier I_2 and with different types of surfaces.

The Pyrex apparatus used in preliminary studies of the volatility of radioiodine will be replaced by similar apparatus made of quartz for study of less volatile materials at higher temperatures and, later, by all-metal apparatus.

¹W. J. Megaw and F. G. May, The Behavior of Iodine Released in Reactor Containers, AERE-R3781 (July 1961).

²J. B. Morris et al., The Removal of Iodine from Air on a Plant Scale, AERE-R3917 (December 1961).

³F. Hudswell et al., The Sorption and Desorption of Iodine, AERE-R3183 (January 1960).

7. CHARACTERIZATION AND CONTROL OF ACCIDENT-RELEASED FISSION PRODUCTS

When radioactive materials appear in gases as gaseous components, or vapors, they can conceivably be removed by adsorbers as is frequently done with iodine and the noble gases. When the radioactive burden of gases takes the form of particles 0.3μ or larger in diameter, it can be removed by high efficiency particulate filters. There is a range of particle sizes, however, much smaller than 0.3μ , where diffusion processes are still too slow for efficient deposition on surfaces and particle masses are too small for efficient filtration by inertial impaction. Particles of this size occur naturally in the atmosphere and may be produced by several processes, including the condensation of dilute vapors, a condition which may prevail in a nuclear accident. These particles are usually of only indirect importance, for example, as condensation nuclei, but may still play an important role in the behavior of radioactive materials, because significant quantities of radioactivity may be associated with

them. Fission products may form small particles directly or deposit on preexisting particles and exhibit apparently anomalous behavior, penetrating both adsorbers and filters. Such behavior has been observed and may be attributed to the presence of fine particles.

To characterize radioactivity carried in gases as to the particle size with which it is associated, measurements based on the radioactivity of the particles are necessary rather than those based on light scattering, condensation of supersaturated vapors, or optical or electron microscopy. The relative effectiveness of particles of different sizes and kinds for carrying radioactivity, as determined by radioactivity measurements, may differ from that indicated by other methods of measurement. Accordingly, an investigation concerned with the transport and deposition of fine particles has recently been undertaken. Also, consideration of the behavior of accompanying vapors such as iodine is included in the study.

PARTICLE SIZE DISTRIBUTION OF RADIOACTIVE AEROSOLS BY DIFFUSION COEFFICIENT MEASUREMENTS

W. E. Browning, Jr.

R. D. Ackley

An important mechanism in aerosol transport and deposition is that of diffusion, and an important characteristic of an aerosol, its particle diameter, is related to the diffusion constant. Townsend¹ measured the diffusion coefficients of ions in gases, and Nolan and Guerrini² and Thomas³ adapted this method to determine the diffusion coefficient for a monodisperse aerosol. The initial emphasis in the present study has been given to the application of diffusion as a means of determining the particle size distribution of aerosols. At the same time, information helpful in understanding the mechanism itself is obtained.

Various investigators have derived expressions for the fraction of the entering particles, assumed uniform and in a flowing gas stream, penetrating rectangular or cylindrical channels. As the equations are rather cumbersome, only the functional relationships will be indicated. For rectangular geometry,

$$N_g/N_0 = f(D, Q, Z, a, b), \quad (1)$$

where N_g = number of particles remaining in the gas, N_0 = number entering, D = their diffusion coefficient, Q = volumetric flow rate, Z = channel length, a = one-half the distance between parallel walls of height b . For a cylindrical tube, the diameter, in effect, cancels itself so that the

¹J. S. Townsend, Phil. Trans. Roy. Soc. London. Ser. A., 193, 129-58 (1900).

²J. J. Nolan and V. H. Guerrini, Proc. Roy. Irish Acad. A., 43, 5-24 (1935).

³J. W. Thomas, The Diffusion Battery Method for Aerosol Particle Size Determination, ORNL-1648 (Dec. 14, 1953).

corresponding equation for N_g/N_0 does not include diameter (or, of course, a or b). However, for the work described here, these equations are not directly applicable since the procedure being employed is to pass an aerosol through channels and then to determine the amount of deposition on definite increments of length of the inside walls of the channel as a function of channel length. Consequently, letting n_s represent the number of particles of a given species per unit length and letting N_s represent the cumulative number, the equations were modified to yield,

$$n_s = \frac{\partial N_s}{\partial Z} = - \frac{\partial N_g}{\partial Z} = -N_0 f_Z(Z) . \quad (2)$$

For a spectrum of particles, having different diffusion coefficients, their distribution on the wall of a channel would be,

$$\sum_i n_{s_i} = - \sum_i N_{0_i} f_Z(D_i, Z) . \quad (3)$$

The actual equations which have been employed are those of DeMarcus⁴ and Gromley and Kennedy⁵ after modification as indicated above. For a rectangular channel, a single species, and a sufficient degree of deposition,

$$-f_Z(Z) = 0.915\beta e^{-\beta Z} + 0.696\beta e^{-11.8\beta Z} + 2.08\beta e^{-80\beta Z} , \quad (4)$$

where

$$\beta = \frac{3.77bD}{aQ} . \quad (5)$$

The corresponding equation for a cylindrical channel is similar to Eq. (4) except that the numerical coefficients are different and now,

$$\beta = \frac{3.66\pi D}{Q} . \quad (6)$$

The approximate diffusion coefficient range of interest, for molecules or particles in air at normal conditions, is from 0.085 cm²/sec for molecular iodine down to approximately 3×10^{-7} cm²/sec for particles 1 μ in diameter. A close examination of Eqs. (4)–(6), along with a consideration of the experimental factors involved, indicates that a cylindrical geometry would be convenient for the smaller particles, but that, for the larger ones, a rectangular geometry would be more useful. In order to facilitate the application of Eq. (3), an IBM nonlinear regression code for processing the data, so as to supply D_i 's and relative N_{0_i} 's, weighted for radioactivity, was utilized.

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

⁵P. G. Gormley and M. Kennedy, Proc. Roy. Irish Acad. A., 52, 163–69 (1949).

The objective of the initial experiments was to verify the theory outlined above. These experiments consisted in passing air through a chamber in which an aerosol was being generated, then through I^{131} vapor, and next through one or two diffusion channels. Radioiodine was used because of its important role in nuclear safety problems and as a means of labeling particles to simulate other fission products. The particles used were formed by slow combustion of tobacco and/or by high frequency discharge (4 Mc, 50 kv) across a spark gap made with aluminum wire. The presumed diameters for these particles are $0.25\ \mu$ for the tobacco smoke, from the value listed by Cadle,⁶ and $0.004\ \mu$ for the aluminum oxide particles obtained by sparking aluminum wire, from a diffusion study by Dearth and Van den Akker,⁷ who used a method of collection and detection very different from that discussed here. The aluminum oxide particles have since been examined, by means of electron microscopy, at 68,000X with the results as shown in Fig. 7.1. Their observed diameters are $0.003\text{--}0.008\ \mu$, representing good agreement with the value $0.004\ \mu$.

A diagram illustrating one of the experiments is shown in Fig. 7.2. In another experiment, in which only the spark gap was employed for particle generation, the rectangular channels were replaced by a single copper tube approximately 60 cm long with an inside diameter of about 0.8 cm. Transport of elemental iodine alone was similarly studied, with a copper tube, by omitting aerosol generation. After exposure of the diffusion channels, the distribution of activity on their walls as a function of length was determined by means of a gamma spectrometer. Some samples of the data which were obtained are shown in Figs. 7.3 and 7.4, along with appropriate theoretical curves. Regarding the experiment corresponding to Fig. 7.3 (and, also Fig. 7.2), the regression analysis of the data from the two channels actually gave a diffusion coefficient equivalent to the aforementioned value of $0.25\ \mu$, although such agreement must be considered partially fortuitous. The deviation between the observed and the calculated values for the 1-cm-spaced channel is associated with low counting rates and, thus, is probably not as serious as it appears. Neither of the two channels showed evidence of elemental iodine or $0.004\ \mu$ particles; this observation is a result of their deposition prior to entering the rectangular diffusion channels. Data in Fig. 7.4 for iodine alone show good agreement with theory, and data for the aluminum oxide particles appear satisfactory except for an entrance effect which may be a result of iodine vapor still being deposited on the particles. It is of interest to note the profound effect of $0.004\ \mu$ particles on deposition of radioiodine. The results of these method evaluation experiments, in which three species with widely differing diffusion coefficients were employed, definitely indicated that this technique might be of considerable value in the characterization of finely divided radioactive particulate matter.

⁶R. D. Cadle, Particle Size Determination, p 1, Interscience, New York (1955).

⁷L. R. Dearth and J. A. Van den Akker, A Study of the Filtration and Permeability Characteristics of IPC 1478 Filter Paper, DASA-1168, pp 37-38 (Feb. 13, 1960).

0.5 μ

Fig. 7.1. Sample of Aluminum Oxide Particles Obtained by Sparking an Aluminum Wire. 68,000X.

Fig. 7.2. Particle Diffusion Experiment with Rectangular Channels.

Fig. 7.3. Diffusional Deposition of Smoke, Labeled with I^{131} , in Rectangular Channels. Particle size $\sim 0.25 \mu$.

Fig. 7.4. Diffusional Deposition of Molecular Iodine and Aluminum Oxide Particles in a Cylindrical Channel.

Among the possible applications of the particle diffusion measurements, one which is currently under investigation involves determining the efficiency with regard to very fine particles of various filtration media such as high-efficiency particulate filters and charcoal beds. The arrangement shown in Fig. 7.5 was evolved after performing several experiments with particles emanating from hot stainless steel ribbon in an air stream; the particles were labeled with I^{131} in the manner indicated in the figure. However, at present, the aluminum oxide particles are again

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Fig. 7.5. Particle Diffusion Apparatus for Investigating the Efficiency of Filters for Removal of Very Fine Particles.

being utilized since their nature and behavior are better defined. This investigation on filters is not sufficiently advanced to warrant firm reporting of efficiencies, but a selected example of the data being obtained is shown in Fig. 7.6 for purposes of illustration. Referring to the inlet data, the steep portion reflects elemental iodine, and the region $Z = 15-45$ cm aluminum oxide particles (with a slightly larger average diameter than observed in Fig. 7.4). Looking at the exit data, one observes that elemental iodine has been almost completely removed while the number of particles was reduced by a factor of approximately 4.

Plans are to continue the filter media efficiency studies, and, also, preparations are being made for extension of the use of this tool to field applications.

Fig. 7.6. Diffusional Deposition from Side-Stream Samples of Air Carrying Al_2O_3 Particles Plus I^{131} Vapor, Before and After Activated Carbon Filtration.

MEASUREMENT OF CHARACTERISTICS OF RADIOACTIVE AEROSOLS USING FIBROUS FILTERS

W. E. Browning, Jr.

M. D. Silverman

An investigation of a method of determining the characteristics of radioactive aerosols using fibrous filters is being undertaken. The initial filters will contain Dacron (staple) fibers, 1-1/2 in. long and 11 μ in diam, which have been carded into a uniform net of material. It is planned to pass air containing radioactive particulates through a series of such filters, with an absolute (CWS) type filter as a final cleanup before venting to the hood exhaust. Radioactive particles (50-500 A) are to be generated by passing a stream of air over a previously irradiated iridium wire heated to 500-1000°C. The radioactive Ir^{192} particles in the air stream will be removed by the series of filters. Distribution of particles through the series of filters will be determined from radioactive counting analyses. The data will be examined with reference to existing theories of filtration and to determine the conditions for removing radioactive particulates from gaseous systems.

PART III CONTAINMENT ENGINEERING

REACTOR DIVISION

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8. NUCLEAR SAFETY PILOT PLANT

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The Nuclear Safety Pilot Plant is an experimental tool for the study of the release and transport of fission products from irradiated fuel, within a model containment vessel, and through a gas-sampling train. In addition, it will allow the investigation of various techniques for the removal of fission products within the containment vessel. Initially, fission products will be released from small samples of preirradiated uranium dioxide fuel, although eventually it is planned to melt down full-scale fuel elements of various fuel and cladding materials.

The design of the facility is well advanced and, except for the heat source, will consist of components and instrumentation which have been proved under comparable service conditions. The heat source, however, was selected for its experimental flexibility to produce melting of potential reactor fuels of interest. A low-momentum plasma torch not only provides this capability but also will permit environmental control with a minimum of extraneous materials. Some development work with the torch has been undertaken.

DESCRIPTION OF THE FACILITY

The Nuclear Safety Pilot Plant (NSPP) facility will consist of a heat source, a furnace, a model containment vessel, a gas-sampling and cleanup system, and a decontamination system. The equipment will be placed in an unused cell in the Homogeneous Reactor Experiment-2 building. The cell is accessible from the top and is lined with stainless steel. The existing 4-ft-thick concrete shielding walls will permit a mixed fission-product inventory in excess of 1000 curies, although it is expected that most experiments will deal with much smaller sources. The facility will be capable of being dismantled and decontaminated remotely but will be assembled by direct procedures.

A proposed flowsheet for the facility is shown in Fig. 8.1. The plasma torch heat source generates an inert gas stream of more than

30,000°F which can be directed on the specimen. In practice, the flame will be diluted to a controlled temperature by inert gas, air, nitrogen, oxygen, or water vapor, depending on the atmosphere desired for the particular experiment. The melting will be performed in a furnace in which a specimen 1 in. in diameter and 30 in. long, supported by a water-cooled boat and a tantalum liner, may be passed under the flame. Initially, most samples will be much smaller than this. Pyrometry and TV viewing will be performed through sight ports. The vaporized material will be swept from the surface of the melt and transferred to the model containment vessel by means of a short heated transfer tube. The released material will either be dumped into the containment vessel or directed to a gas-sampling train, depending on the particular experiment.

The model containment vessel is a vertical cylindrical stainless steel tank 10 ft in diameter and 15 ft long. The vessel will be instrumented to determine the disposition of the fission products and associated debris by use of paper tape air samplers, metal coupons, and fallout trays. Also, means will be available for testing countermeasures such as recycle filtration of the containment vessel atmosphere, water spray, and combined steam and water spray removal of small particles by using their properties as condensation nuclei. A decontamination system is built into the vessel for cleanup and total inventory sampling. Liquid wastes will be transferred to an existing concrete decontamination pit for sampling and will be disposed through existing facilities at the HRT site. The interior of the vessel will be accessible through a 36-in. port at the top and an 18-in. manhole in the side. In addition, two 8-in. ports and two 8-in. purge flow ports will be available in various combinations for added flexibility. The vessel will be designed to withstand 15-psig external pressure and approximately 50-psig internal pressure.

Two gas-sampling trains will be used initially, one capable of sampling 1% of the containment vessel volume per day, the other sized to handle the normal torch flow. The low-flow system will be used to evaluate the effects of leakage from the vessel at controlled rates through controlled cracks or openings. The high-flow system will be used primarily to determine the composition of the material released from the fuel. As shown in Fig. 8.1, the sampling trains consist of Millipore filters, iodine traps (either charcoal or silver-plated metal mesh), dehumidifying traps, and charcoal xenon and krypton traps operating at dry-ice temperature. Fractionation of the rare gases can be effected by utilizing the different breakthrough times in charcoal at dry-ice temperature. This technique will alleviate the problem of oxygen condensation on the charcoal with its attendant explosion hazard that would exist if liquid nitrogen were used for the krypton-trap coolant.

The equipment will be placed in the HRT B cell as shown in Fig. 8.2. The gas-cleanup systems will be arranged so that the various traps and filters can be removed remotely by means of long-handled tools operated through holes in the top shield. All filters, traps, and piping will be connected by Grayloc fittings that can be disconnected remotely. Piping runs will be of lengths such that sections can be removed remotely and decontaminated. It appears that viewing will be done either by periscope or closed-circuit TV. The design does not preclude the future use of manipulators if their use is found to be worthwhile.

Fig. 8.2. Nuclear Safety Pilot Plant Equipment Layout.

Secondary containment for the plant will be effected by maintaining the cell at subatmospheric pressure using the existing HRT stack-house blowers. Removal of samples from the containment vessel prior to decontamination will be done with long-handled tools through small openings. Sufficient air flow will be maintained into the openings by the stack blowers, through ducting to the containment vessel, to prevent back diffusion of harmful material. This flow will be passed through particulate filters and iodine-removal beds prior to exhaust up the stack. The rate of flow through this purge system will be controlled by the allowable concentration of radioactive nuclides in the stack exhaust. The model containment vessel will be purged at increasing rates until sufficient flow exists to provide 100 ft/min inward air flow through the openings used. A purge flow of 1000 scfm is deemed sufficient to do this. Booster blowers will be used in addition to the stack-house blowers to provide the necessary flow and standby capability. The existing stack filters are considered as backup for the filters inside the cell; under usual circumstances, excessive radioactive material will not leave the boundaries of cell B.

THE HEAT SOURCE

The heat sources considered for the NSPP were induction heating, resistance heating of the sample, resistance heating of a heater with radiant heating of the sample, and heating by means of a plasma torch. Induction heating was found to be too unwieldy and expensive in the power sizes and frequency ranges required for this application. Resistance heating of the sample would require considerable development because of the poor electrical conductivity of UO_2 at low temperatures and the difficulty of maintaining power input once melting had begun. Radiant heating from a resistance heater would be too limiting because of the requirement of low heat loss and the fact that graphite would be the only feasible heater element and would preclude the use of oxidizing atmospheres during the experiments.

A plasma torch was chosen as the heat source because of the intense temperatures and power inputs that could be imparted to the sample, the purity that could be attained for the gas stream playing on the sample, the ability of providing practically any gaseous atmosphere around the sample by judicious dilution of the primary stream, the small size and flexibility of the torch itself, the lack of restriction on the length of electrical leads from the power supply to the torch, and the fact that torch systems are available as proved off-the-shelf items requiring very short delivery lead times.

Torches emanating a turbulent gas flame would be a problem in that the extremely friable UO_2 specimens could be blown out of the heating area without significant melting. Recent advances in torch design allowing a laminar low-momentum flame have solved this problem. Also, recent torch designs allow the sample to act as an electrode after being heated to a conducting temperature by the normal mode of operation. By this means, known as transfer arc operation, volume heating can be effected in the sample. The spallation and splattering effects of early plasma torches are almost nonexistent with the models now available.

EXPERIMENTAL WORK ON PLASMA TORCH

Tests have been performed to determine (1) that a UO_2 -melt holder could be developed, (2) the range of particle sizes collected from the vapor onto a Millipore filter, and (3) the fraction of UO_2 vaporized from the melt.

The initial tests were performed in equipment which was borrowed from another laboratory group. An EGCR natural- UO_2 pellet was contained in a tantalum crucible, and the tantalum crucible held in a water-cooled copper block as shown in Fig. 8.3. A layer of zirconia powder was provided between the tantalum and the copper block to effect a small amount of insulation. The plasma torch was held so that the flame impinged on the UO_2 . The UO_2 cracked from thermal shock, and the pieces melted with no difficulty in less than 1 min. The tantalum holder was partially melted. The copper crucible holder showed no signs of damage after this run and was

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Fig. 8.3. UO_2 Melt Holder.

used in all of the experiments conducted without showing sign of damage. It was concluded from such preliminary tests that a water-cooled copper block could be used to support the uranium fuel and its primary container during the melting process.

A series of tests was then conducted to determine the range of particle sizes of the vaporized UO_2 . In these tests the UO_2 slugs were melted as before, but the off-gas was passed through a type VF Millipore filter, and electron micrographs were made of the dust collected. PWR-type slugs of UO_2 weighing about 15 g were melted in the enclosed stainless steel container shown schematically in Fig. 8.4. The off-gas flow of 10 scfh was passed through a water-cooled tube (0.87 in. OD by 18 in. long), a 4-ft length of 1-in.-ID Tygon tube, and collected on the type VF Millipore filter. The Millipore filter was submitted for particle-size and chemical analysis. In addition, particulate material was removed from the furnace walls and analyzed for uranium.

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Fig. 8.4. Schematic of Particle Collector for UO_2 Smoke.

The range of particle sizes of UO_2 as measured from the electron micrograph is from 0.01 to 0.4 μ . The larger sizes were agglomerates of the small particles. An electron diffraction measurement showed the average crystallite size (of U_3O_8) to be about 0.04 to 0.05 μ . An electron micrograph of a carbon replica of a portion of a Millipore filter (Run 1) is shown in Fig. 8.5. The magnification is 50,000X. The lacy structure is the replica of the filter surface. An electron micrograph of the dust collected from the surface of the furnace is shown in Fig. 8.6. The particle size range is 0.005 to 0.2 μ .

Fig. 8.5. Electron Micrograph of Millipore Filter. Particle size range 0.01 to 0.4 μ .

Fig. 8.6. Electron Micrograph of Duct Particles Collected from Furnace Wall. Particle size range 0.005 to 0.20 μ .

The above melts were used to determine the fraction of the UO_2 vaporized during melting. The initial UO_2 slug in each experiment weighed between 14 and 15 g. The amount of uranium left in the melt was determined by chemical analysis after the run. The difference was taken to be the amount of UO_2 equivalent that was vaporized. The conditions of the run and the results in terms of percent vaporized and percent vaporization rate are summarized in Table 8.1. The vaporization rates varied from 0.2164 %/min at 12.5 kw to 3.46 %/min at 20 kw of plasma power. It is clear from examination of Table 8.1 that the vaporization rate of UO_2 depends upon the power input to the plasma torch and that the total amount vaporized depends upon the power input and the exposure. This, of course, is to be expected.

Table 8.1. Plasma-Torch Vaporization of PWR-Type UO_2 Slugs

Run No.	Plasma Power (kw)	Time of Exposure (min)	Gas Flow (scfh)	Initial Weight of UO_2 (g)	UO_2 Vaporized		
					Grams	Percent	Percent/min
1	20	3	10	14.285	1.484	10.39	3.46
2	18.75	1.5	10	14.538	0.535	3.68	2.45
3	12.5	5.0	10	14.611	0.158	1.082	0.2164

These data show that the rate of activity release due to vaporization can be controlled by using the plasma torch. This will make it possible to study the decontamination factors within the NSPP under different activity input rates.

A plasma torch for use in the NSPP was recently received and is now being used to perfect the zone-melting techniques. Studies are also being made on the loading and unloading techniques which will be employed for the irradiated UO_2 fuel slugs in the NSPP.

PROJECTED EXPERIMENTAL PROGRAM

Although potentially capable of being used for investigation of a wide range of factors affecting the release and transport of fission products, the facility will be used initially to find answers to what at this time appear to be some of the most urgent questions relating to reactor safety. These questions involve the release of biologically important fission products from molten irradiated fuel, the transport properties within the containment vessel, the release from leakages in the containment vessel, and the effectiveness of countermeasures that may be taken to reduce the amount of radioactivity reaching the environment. With the above questions in mind, the following procedure has been mapped out.

The facility will be operated initially with nonirradiated material to check the performance of the integrated system (torch and furnace development will be concurrent with the construction of the rest of the facility). This should check the operability of the system and, in addition, yield some data on particle size distribution in various parts of the system.

Following this, low-level long-cooled bare UO_2 will be melted to determine the disposition of the fewer longer-lived fission products. This will allow operation of the facility under much less complicated conditions than would exist with a full spectrum of fission products, and, if difficulties should arise, the activity levels should be low enough to permit direct maintenance. This will also check the decontamination and liquid-sampling systems.

If the above can be done successfully, small samples of short-cooled UO_2 of from 1 to 100 curies will be melted to determine the disposition of the shorter-lived fission products such as the iodines. These meltdowns will be done in atmospheres of helium, air, and steam to determine the effects of these atmospheres and to compare results with laboratory-scale experiments already performed.

Since the evaluation of behavior in containment vessels is of prime importance, these tests will be begun as quickly as characterization of the release from the melts can be done. The first containment vessel tests will be performed with no countermeasures. In this series, the disposition of fission products as a function of time and position in the vessel will be determined by means of air samplers, fallout trays, coupons of various materials, and air current measurements. The effects of recycling the containment vessel atmosphere over particulate and iodine filters will be assessed. Studies will also be made with various gas flow rates, times from particulate release to initiation of the recycle operation to determine effects of changes in the chemical character of iodine with time, and length of times of operation. Further tests will be run in which attempts will be made to remove the fission products from the model containment vessel by using the condensation nuclei properties of fine particulate matter. These tests will be performed by introducing steam into the containment vessel followed by a quenching water spray. The effects of water spray alone will also be studied. These experiments will also serve to evaluate laboratory data on a larger scale as a step toward the application of design data to full-size nuclear power plant conditions. As part of this phase of the program, containment vessel leakage studies will be performed in which the vessel will be pressurized, with air or steam, and allowed to leak at a rate of approximately 1% of the total volume per day through small controlled leaks into the low-flow gas-sampling system.

Further meltdowns will be performed varying parameters that were not previously investigated. For instance, UO_2 may be melted with stainless steel or Zircaloy cladding, perhaps using full size fuel elements and varying the atmosphere under which meltdown takes place. Fuels other than UO_2 can be heated, such as metallic elements, carbide fuels, and graphite-uranium carbide dispersion elements.

Concurrent with NSPP tests, work will be progressing on correlating data obtained in laboratory-scale out-of-pile melting, in-pile melting, and the planned SPERT destructive tests. If correlations can be found between the various means of heating, the NSPP will be useful in studying the release of fission products from fuel in detail in addition to providing a test bed for the investigation of these products after release. If these correlations can be made, the ability of the facility to heat full-scale fuel elements will be invaluable for release tests below melting temperatures as well as in the melting and vaporization range.

Radiochemical analyses will be performed with existing facilities at ORNL. If, however, on-site gamma spectroscopy proves feasible, this will be done as much as possible, where applicable. It is estimated that approximately one experiment per month will be feasible. Under this assumption, three years will be required to evaluate effectively containment vessel effects and countermeasures and to characterize the release in various atmospheres. Also, the above program may be altered considerably depending on the urgency of new questions that arise and unforeseen changes in the capability of the facility.

PRESENT STATUS OF DESIGN WORK

At this date the configuration of the plant for the first series of experiments has been decided. The process flowsheets are complete, and instrumentation flowsheets are nearing completion. The model containment vessel has been designed and let out for bids, and bids have been received on a plasma torch capable of a laminar flame and transfer arc operation with a 50-kw capability. The cell layout is essentially complete. The preliminary design of the furnace has been evolved, and construction drawings have been started. Drawings of various filter holders and traps are under way, and the design of the recycle filter system has started.

It is estimated that the NSPP expenditures will total \$555,000 at the end of FY 1963. This will cover costs of the development of the furnace; the design, purchase, and installation of all components in the B cell; and preliminary runs with unirradiated UO_2 elements. It is anticipated that the test runs with unirradiated UO_2 elements will start March 1, 1963. Tests with long-cooled UO_2 elements will start about July 1, 1963. The elements for the latter tests are available.

9. REACTOR CONTAINMENT HANDBOOK

C. L. Whitmarsh

A handbook on the subject of reactor containment is being prepared by Oak Ridge National Laboratory. This document will provide sufficient information, in the form of analytical techniques, process descriptions,

and experimental data, for the conceptual design of a containment system for a reactor. Most of the work will be performed by ORNL personnel although selected portions of the handbook will be prepared by an architect-engineer firm active in the containment construction field. The handbook is scheduled for completion in 1963.

The handbook will serve as a working guide for the containment-design engineer. Procedures will be suggested for designing the various types of containment systems - high and low pressure, pressure suppression, pressure relief, negative pressure, etc. - under consideration in the nuclear reactor industry. However, these procedures will be sufficiently flexible to allow the designer to utilize new data and original methods. Background information will include analytical treatments of the fundamental processes involved and the data and experience pertinent to these processes. Credible accidents which affect the containment system will be covered in the analysis.

A tentative outline and an estimate of the status of the first draft is given below.

REACTOR CONTAINMENT HANDBOOK

1. INTRODUCTION (> 50% complete)
 - Philosophy and Purpose of Containment
 - Reactor Types (general description)
 - Containment Systems (general description)
2. CODES, CRITERIA AND REGULATIONS (< 50% complete)
 - AEC Regulations
 - Regulating Ordinances of Other Agencies
 - Technical Codes and Criteria
 - Design Safety Factors
 - Insurance Requirements
3. RADIOACTIVITY - GENERATION AND TRANSPORT (< 50% complete)
 - Generation and Release
 - Transport Within Primary System
 - Transport Within the Outer Containment
 - Transport External to All Containment Barriers
 - Washdown, Spray, and Other Nonnatural Techniques for Reducing Amount of Fission Product Transport
4. ENERGY SOURCES (completed)
 - Nuclear
 - Chemical Reactions
 - Latent Heat and Stored Energy

5. ANALYTICAL TECHNIQUES (< 50% complete)
 - Heat Transfer Problems
 - P-V-T Relationships
 - Hydraulic Problems
 - Structural
 - Shock and Missile Protection
 - External Loading
 - Computer Programs
6. CREDIBLE ACCIDENTS (not started)
 - Accidents as a Function of Reactor Type
 - Consequence of Energy Release in Maximum Credible Accident
 - Miscellaneous Accidents
 - Use and Effect of Pressure Reduction Schemes, External and Internal Sprays, Condensing Pools, etc.
7. SPECIFIC CONTAINMENT SYSTEMS (not started)
 - High-Pressure Containment
 - Low-Pressure Containment
 - Pressure Suppression
 - Pressure Venting
 - Negative Pressure (confinement)
 - Others
8. CONTAINMENT PROOF TESTS (not started)
 - Significant Leak Paths
 - Leakage of Various Contents
 - Structural Integrity
 - Gross-Leakage Tests
 - Localized Leakage
 - Maintenance of Containment
9. MATERIALS CONSIDERATIONS FOR USE IN ANALYSIS (not started)
 - Lists of Materials and Use
 - Heat Capacities and Transfer Coefficients
 - Testing Methods, for Materials Properties, and Limitations
 - Static Properties - Normal Test Conditions
 - Dynamic and Transient Response Properties - Normal Test Conditions
 - Effect of Operating Conditions
 - Fatigue Limits
 - Statistical Analysis of Available Data on Little-Known Properties
 - Correlating and Extrapolating from Easily Obtained Properties
 - Shop Processes - Inspections and Requirements Necessary to Obtain Desired Properties
10. CONSTRUCTION DETAILS AND ACCESSORIES (not started)
 - Design Principles and Specifications
 - Fabrication Techniques, Workmanship, Inspection
 - Airlocks, Hatches, Service Connections
 - Valves, Exhaust Systems, Ducts
 - Sealing Methods
 - Support Structures

11. ECONOMICS (not started)

- Effect on Total Power Costs

- Containment System Cost vs Location, Material of Construction,
Multiple Vessel Arrangement, Geometry, etc.

- Future Outlook for Containment Systems

12. CONTAINMENT RESEARCH (not started)

- Programs in Progress - Governmental and Private Industry

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PART IV RADIOCHEMICAL PLANTS

CHEMICAL TECHNOLOGY DIVISION

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10. RADIOCHEMICAL PLANT SAFETY STUDIES

C. E. Guthrie

E. D. Arnold

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 an adequate understanding and assessment of the hazards and means of combating them be available before such a plant is built. In order to accomplish this three general areas must be covered. These are (1) assessing the credible radiochemical plant accidents, 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.

An outline of a radiochemical plant hazards analysis is shown in Table 10.1. Calculations are being made to determine the maximum downwind hazard due to both contamination and personnel exposure from each of the accidents as a function of the degree of containment, degree of dispersal of contaminants, aerosol properties and meteorological conditions. The evaluation is based on hazards analysis calculation methods developed to evaluate ORNL facilities during the containment program.¹ The calculation methods are being expanded and refined in order to cover the extended scope of this study. This will be followed by a more detailed study of those factors which prove to be the most significant in contributing to or increasing the hazard as a result of an accident.

¹E. D. Arnold and J. P. Nichols, "Hazards Analysis for Chemical Processing Facilities," Nuclear Safety 3(1), 56 (1961).

Table 10.1. Hazards Survey Outline

Facility criteria: Fuel processed, 1 ton/day Yankee
 Source fabrication, 10,000 curie Sr^{90}
 Cell volume, 8000 ft^3
 Building volume, 300,000 ft^3
 Cell ventilation rate, 800 cfm
 Cell leak rate, variable
 Stack dilution factor, $1.6 \times 10^{-5} \text{ sec/m}^3$

 Accidents To Be Investigated

1. Waste evaporator chemical explosion
 - a. Filters ruptured
 - b. Filters not ruptured
 2. Vessel rupture in Pu extraction system due to criticality incident of 10^{19} fissions
 - a. Filters ruptured, no I_2 absorbers
 - b. Filters not ruptured, no I_2 absorbers
 - c. I_2 absorbers
 3. Criticality incident in Pu product handling (10^{17} fissions)
 - a. Glove box rupture
 - b. Glove box intact
 4. Chemical explosion in 10,000 curie Sr^{90} source preparation
 - a. Wet system
 - b. Dry system
-

It is apparent from the work to date, and past radiochemical plant experience, that the absolute filters now in use are damaged by chemical fumes and moisture both of which are normally present in hood, vessel, and cell ventilation. A relatively minor accident in a cell, such as a steam line break, could cause the absolute filters to rupture and release any retained activity as well as activity in the exhaust at that time. A preliminary investigation has been made to find a material which would be more suitable than the paper, asbestos, or glass fiber now in use for radiochemical plant absolute filter material. Graphite fibers appear to be the most promising candidate. Graphite fibers are chemically resistant, do not oxidize in air up to $\sim 700^\circ\text{F}$, and, since they are not wet by water, should not plug and lose strength in moist air. The technology already exists for production of graphite fibers in the size range required ($\sim 3 \mu$ diam). Preliminary contacts with filter manufacturers have been made in order to establish a program for development of a graphite absolute filter.

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