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EXPERIMENTS ON THE RELEASE OF FISSION
PRODUCTS FROM MOLTEN REACTOR FUELS

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EXPERIMENTS ON THE RELEASE OF FISSION PRODUCTS FROM MOLTEN REACTOR FUELS

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ABSTRACT

Experiments on the release of fission products in the controlled melting of irradiated fuel of the APPR, STR, and MTR types under conditions simulating the loss-of-coolant accident have been performed. The results indicate that a moderate amount of heating in air or steam sufficient only to melt a specimen results in the partial volatilization of the rare gases, the halogens, and the alkali metals. In the presence of air or steam, little of the strontium and other fission products is released. In this type of experiment the only particulate activity observed was composed entirely of cesium.

At the level of trace concentration of fission products, APPR plate melted at 1525°C and released 50% of the rare gases, 33% of the iodine, 9% of the cesium, and traces of strontium. After 25% burnup the cesium value rose markedly to 41%, while the iodine value apparently declined to 12%. Aluminum alloy at 700°C released only 2% of the iodine and 10% of the rare gases. Zirconium alloy, after 15% burnup, melted at 1850°C in air and released all of the rare gases, 26% of the iodine, 10% of the cesium, and less than 1% of the strontium. When melted in steam, approximately 2% of the strontium and 0.5% of the barium were also released from zirconium alloy.

Exploratory experiments have been performed with Geneva-type aluminum-dispersion fuel, UO₂, and GE-HTRE fuel.

INTRODUCTION

In considering the hazards associated with the operation of nuclear reactors it has been necessary to assume that gross nuclear accidents always involve the release to the atmosphere of the entire radioactive contents of the reactor. This assumption was made since no valid data existed on the extent of fission product release when a reactor core was destroyed. A serious consequence of this unfortunate assumption is that a large amount of land must be designated as an exclusion area around a reactor site in order to ensure the safety of the adjacent population. This area is a function of the size of the reactor, and it has already been noted that in some eastern states there are no acceptable sites on which to build a large power reactor because of the immense tract of land required. It is also certain that local or state laws which would seriously impair the prospects of nuclear power are being considered in this light. It is apparent then that any investigation which does establish that, in fact, only a fraction of this amount of radioactive material is

likely to be released will substantially lessen the economic burden on the development of nuclear power. The magnitude of the economic significance of a "major release" as compared with that of a "volatile release" already has been estimated.¹

Some preliminary results of these experiments have been reported previously.²⁻⁴ The present report may be considered as both a summary of the previous papers and a review of the latest results of a similar series of experiments. The work on fission product release is part of the broad program

¹*Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants*, WASH-704 (March 1957).

²G. W. Parker and G. E. Creek, *The Volatilization of Fission Products by Melting of Reactor Fuel Plates*, ORNL CF-57-6-87 (July 15, 1957).

³*Reactor Safety Conference, New York City, October 31, 1957*, TID-7549, Pt. 2 (May 15, 1958).

⁴G. W. Parker and G. E. Creek, *Second U.N. Intern. Conf. Peaceful Uses Atomic Energy, Geneva, 1958*, paper A/Conf. 15/P/1074, United Nations, Geneva, 1958.

sponsored by the Reactor Safety Section of the Division of Reactor Development, AEC. Other phases of the reactor safety program include studies in reactor kinetics characterized by the SPERT, KEWB, and TREAT projects, the metal-water reaction, reactor containment, and development of "reactor fuses."

The Loss-of-Coolant Accident and Its Bearing on the Fuel Melting Experimental Program

In seeking a natural basis for conducting melting experiments, the loss-of-coolant accident, which can be postulated for most reactors and which tends to limit the meltdown process to a finite number of variations, such as the time-to-melt, has been used as the principal guide for experimentation. An estimate of this time interval, which is controlled by the residual gamma-ray heating, can be calculated in terms of power density. In all cases the time to reach maximum temperature is short (several seconds to several minutes). In a few instances, for example, the APPR-1A, the melting temperature may not be reached at all since the maximum should occur at perhaps 1300°C. A range in heating times within the limits of the loss-of-coolant process has been recognized as one of the major areas to be examined in detail.

Partial melting, such as may be induced by a power excursion, should occur in shorter times than the loss-of-coolant process, and this has been simulated by a very fast melting requiring only a few seconds.

The Basis for Threshold Melting

In the melting experiments accomplished to date, only very small specimens have been used. At this scale it is likely that little significant temperature gradient exists throughout the sample and that melting is completed almost instantly. Since it is reasoned that once molten, in the fluid state, the fuel-bearing metal will flow out of the heat zone to an extent depending on its viscosity and containment, it has become a practice to perform only "threshold" melting. By this, it is meant that the total time for which a sample is molten has been kept short. In practice it is limited only by the time required for the visual reflexes of the operator, who observes the process with an optical pyrometer. The cooling of the melt is extremely rapid since little material other than the sample itself is heated.

Objective of the Experiments

In the few historic nuclear accidents experienced in the recent past (e.g., the accidents to the NRX reactor at Chalk River and the EBR-1 and the more recent uranium fire in the Windscale No. 1 reactor) the disaster involved the partial meltdown or overheating and actual combustion of fuel.

In these cases at the time of the incident, information was lacking about the extent of fission product vaporization to be expected, and hence no prediction could be made of the apparent biological hazard created until long after the time of the incident. An evaluation of the anticipated total biological hazard would have been facilitated by a minimum knowledge of the nature and magnitude of the fission product activity which could be expected to be released to the environment in the vapor cloud and to a lesser degree by information on particle size distribution.

Fuel Melting Program

The immediate goal of this experimental program was an early estimate of the percentages of the various fission elements vaporized or entrained in fission product release initiated by a quiet meltdown of various typical reactor fuels. As time permitted it was possible to consider further the effect of the environment, that is, the atmosphere, whether air, inert gas, steam, or water. The study of the metal-water reaction was to be considered only as it affected the release of fission products. An obvious question to be answered was the effect of trace vs macro concentration of fission products.

The work as envisioned was to be conducted in "miniature," that is, at less than fuel-element size. Experiments at full fuel-element scale are being conducted by others. For example, MSA Research Corporation,⁵ Callery, Pennsylvania, in cooperation with the Westinghouse Atomic Power Division is engaged in a program of melting zirconium-alloy fuel elements in a steam atmosphere with provisions to determine the range of particles released and the extent of the metal-water reaction. Under a joint USAF-AEC program

⁵*Fission Product Release During a Simulated Meltdown of a PWR Type Core*, MSA Research Corp., Technical Report 63 (October 1958).

Convair,⁶ Division of General Dynamics, Fort Worth, is conducting field tests on the diffusion and deposition of fission products from ground-level release using melted reactor fuel elements as a source. At Hanford the General Electric Company⁷ is conducting tests on uranium oxidation and simultaneous release of fission products in a manner somewhat comparable to the Windscale Reactor incident of October 1957.

Other reactor fuels were to be considered as they appeared and to an extent in proportion to their usage. In order to enter the experimental program as quickly as possible and at the widest scope it was most convenient to begin by using samples cut or die-punched from typical plates. As the program progressed, specially fabricated miniature fuel elements were made available in some cases (APPR and STR) so that any effect of the exposure of a fuel-bearing layer in an edge would become apparent.

An illustration of some types of fuel plates, disks, and miniatures is seen in Fig. 1.

THEORETICAL CONSIDERATIONS

Fortunately, the widespread interest in the pyrometallurgy⁸⁻¹² of uranium has disclosed a systematic behavior of the fission elements. From this work and especially from the early theoretical work of Brewer¹³ it is possible to predict that a fission product may be released or retained in the

solid residue of a melt according to the following pattern:

Processes favoring release to environment

1. Vaporization of the elemental form of a volatile constituent; this is particularly important for elements such as the rare gases, Xe and Kr, for I₂ and Br₂, and for Cs and Rb
2. Diffusion into an oxide, slag, or skin followed by dispersion or vaporization of the oxide at high temperature; this may occur with Mo and Tc
3. High-temperature vaporization of the element in the absence of air (e.g., Sr and Ba)
4. Formation of chemical compounds between two fission products, particularly with halides
5. Turbulence or purging in the molten state, which induces an increase in apparent diffusion rate

Processes for retention by the melt

1. Occlusion of elemental or combined forms of volatile constituents; the rare gases Xe and Kr are held by molten aluminum unless purged
2. Conversion to nonvolatile carbides (e.g., Zr and Nb)
3. Occlusion in crystalline fuel (e.g., in UO₂ in a molten stainless steel matrix)
4. Formation of stable alloys with the uranium fuel (e.g., Te and Ru)
5. Formation of nonvolatile oxides (e.g., Ba and Sr)

In order to evaluate the behavior of individual fission elements, it is of value to refer to the thermodynamic stability of the oxides as given by Glassner¹⁴ (Table 1). The significance of the free energy of formation is particularly noteworthy in the low values attributed to the oxides of Ru, Cs, Te, and Mo. Since cesium and tellurium are both volatile and both readily dissociated as oxides, equal release from molten uranium would be expected; however, tellurium, as well as ruthenium and molybdenum, appears to have alloying properties which interfere with its escape.

The vapor pressure data given in Fig. 2 (ref 15) may be used in predicting the behavior of the volatile elements cesium and rubidium, as well as barium and strontium. The presence of air effectively prevents vaporization of the alkaline-earth metals barium and strontium, since their

⁶Fission Product Field Release Tests - Experiment Design, FZM-1707 (June 6, 1958).

⁷R. K. Hillard, HW-52753 (Nov. 1, 1957) (classified).

⁸F. R. Bruce et al. (eds.), *Progress in Nuclear Energy, Process Chemistry*, chap. 7, McGraw-Hill, New York, 1956.

⁹F. H. Spedding et al., *Removal of Fission Products from Molten Uranium by Diffusion and Chemical Reaction*, MUC-NS-3068 (1942).

¹⁰H. M. Feder, N. R. Chellew, and M. Ader, *Purification of Nuclear Fuels by Melting in Refractory Oxide Crucibles*, AM-5255 (March 1954).

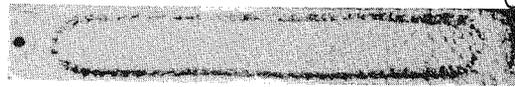
¹¹A. M. Saul, *Pyrochemical Separations Methods: III. The Removal of Fission Products from Molten, Irradiated Uranium by Solid Oxides*, NAA-SR-1361 (Sept. 1, 1955).

¹²F. J. Keneshea, Jr., A. M. Saul, and C. T. Young, *The Distribution of Tracer Plutonium and Fission Products Between Molten Uranium and Solid Uranium Oxide, Carbide and Nitride*, NAA-SR-1032 (Sept. 15, 1954).

¹³L. Brewer, *High Temperature Decontamination and Separation Processes*, UCRL-314 (May 6, 1949).

¹⁴A. Glassner, *A Survey of the Free Energies of Formation of the Fluorides, Chlorides, and Oxides of the Elements to 2500°K*, ANL-5107 (August 1953).

¹⁵R. L. Loftness, *A Vapor Pressure Chart for Metals*, NAA-SR-132 (July 10, 1952).



ZIRCONIUM ALLOY
(STR)



Uncoated



Coated

STAINLESS STEEL-UO₂
(APPR)



Zirconium Capsule
UO₂ Pellet

(PWR)



Stainless Capsule
UO₂ Pellet

(ORGCR)

MINIATURE FUEL SPECIMENS



Stainless-UO₂
(APPR)



Aluminum-UO₂
(Geneva)



Aluminum Alloy
(MTR)



Zirconium Alloy
(STR)



Niobium Alloy
(Experimental)

DIE PUNCHED SPECIMENS

Fig. 1. Typical Fuel Samples for Melting Experiments.

Table 1. Free Energy of Formation* of Oxides of Fission Elements**

Compound	1000°K	1500°K	2000°K
La ₂ O ₃	123	112	100
Ce ₂ O ₃	120	107	94
Pr ₂ O ₃	120	107	94
SrO	116	102	80
UO ₂	109	99	88
ZrO ₂	108	97	86
BaO	111	95	80
PuO ₂	100	95	85
Nb ₂ O ₅	76	66	61
MoO ₂	46	39	32
TeO ₂	16	5	-7.3
Cs ₂ O	35	3.6	-16
RuO ₂	6	-2.7	-10

*Kilogram calories per mole.

**A. Glassner, *A Survey of the Free Energies of Formation of the Fluorides, Chlorides, and Oxides of the Elements to 2500°K*, ANL-5107 (August 1953).

oxides do not dissociate readily. Upon melting in a vacuum¹⁶ or a noble gas atmosphere, these two elements show increased volatility.

FUEL MELTING EQUIPMENT

The heating equipment used was of conventional design and variety. For the low-melting aluminum-base fuels, a simple resistance furnace was suitable and permitted sustained heating and control at the melting temperature. The higher temperatures (to 1550°C) in the resistance furnace (Fig. 3) were obtained by using a platinum winding with a special Mullite combustion tube.

For most higher-melting substances, as well as for quick heating, the induction method was used, assisted by a graphite susceptor ring since most specimens were too small for direct coupling with the high-frequency power. Such equipment is shown in Fig. 4. The power unit itself is not

shown; the characteristics of this unit were a maximum power output of 20 kw and a frequency of 220 kc. For induction melting, the graphite heat source was placed near the central portion of the long silica furnace tube, with a heat-conserving reflector of Al₂O₃ around it. At the melting point of stainless steel (1500°C) little damage was suffered by the silica tube; however, in melting Zircaloy (1850°C) some compromise in duration of heating time was necessary in order to melt the metal before the silica tube was destroyed. More recently it was found possible to accomplish sufficient direct r-f induction with small metal capsules used as a container for die-punched samples.

In order to permit studying the effect of the environment, the atmosphere in the furnace tube could be controlled. The fuel sample was contained in a silica capsule (Fig. 5) suspended by means of a tantalum or platinum wire or a silica rod and was observed with an optical pyrometer until it melted. In some experiments the specimen was loaded and held in other ways, such as in contact with a flat disk of ZrO₂ or by suspension from a metal wire.

PROCEDURE AND ANALYTICAL METHOD

The procedure involved setting up the necessary absorbents, cold traps, and filters and leaving the furnace tube open. Several samples were monitored for comparative total initial gamma activity with an ionization chamber (to allow for accurate comparison) and a vibrating-reed electrometer (Fig. 6) and then were introduced individually in a silica crucible into the furnace tube, to which the filter and adsorbent train were attached. A suitable gas flow (0.3 liter/min) was sustained by means of a vacuum line and supply lines if helium or steam was being used. The furnace tube and specimen were arranged so that an optical pyrometer could be used to observe the heating and the actual melting temperature. Heating was continued only long enough to accomplish melting, which occurred in about 10 to 90 sec, varying for different materials. The melt residue in the small crucible was dissolved in aqua regia or other suitable solvent or placed in solution by fusion, with the necessary provisions for retention of the halogens and the rare gases evolved. All dispersed activity outside the crucible was included in the washings

¹⁶C. T. Young and C. A. Smith, *Preliminary Experiments on Fission Product Diffusion from Uranium-Impregnated Graphite in the Range 1800°-2200°C*, NAA-SR-232 (March 25, 1953).

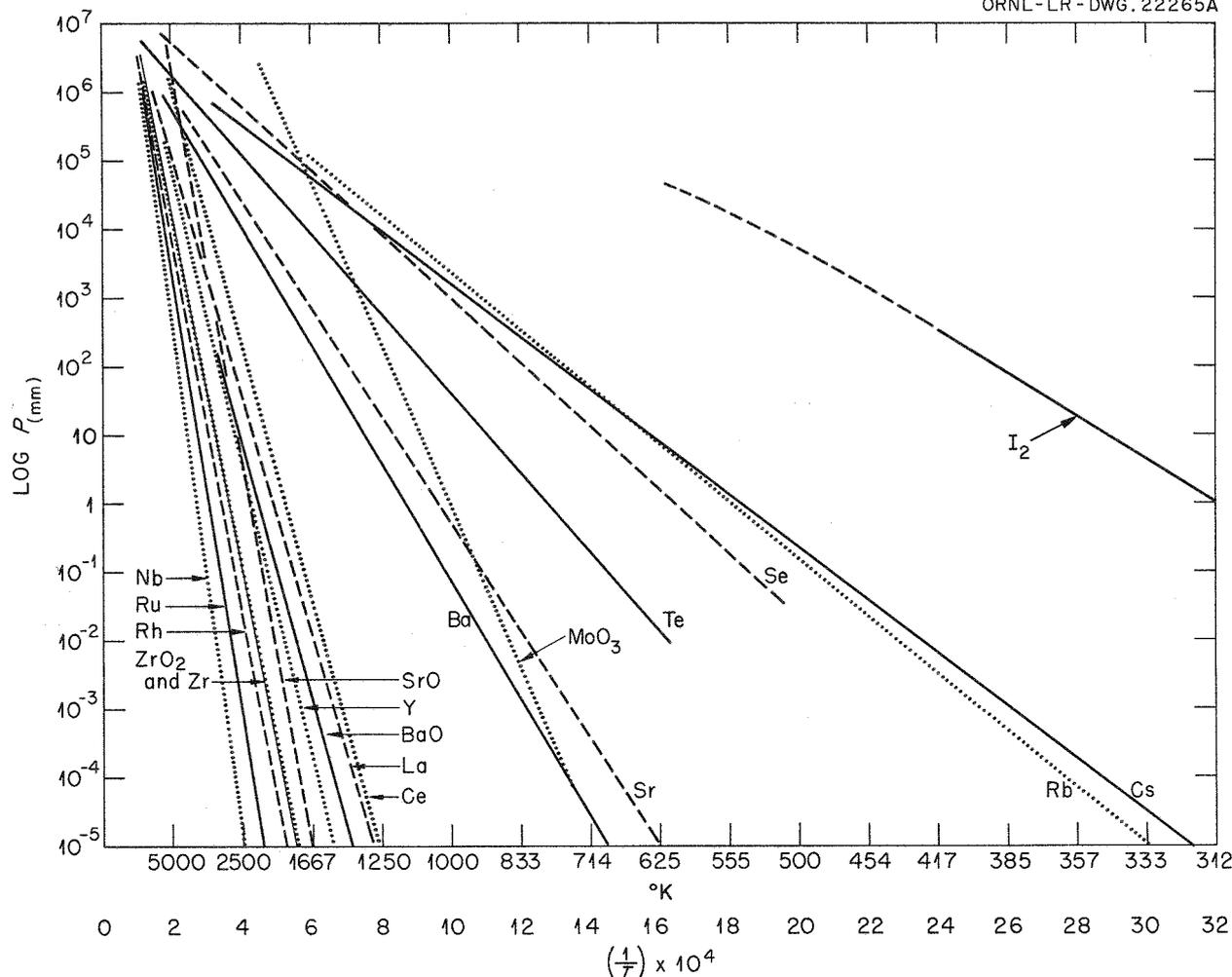


Fig. 2. Vapor Pressures of Metals and Oxides.

from the furnace tube. This washing was accomplished by using the iodine absorbent (NaOH pellets) with water as an alkaline wash and was followed by a rinse with nitric acid and ammonium fluoride.

The volatile products of the melting operation were trapped from the effluent gas selectively either on a filter, by a chemical absorbent in the case of iodine, or on activated charcoal for the rare gases.

The dispersed solids or condensable vapors were collected on a two-stage filter generally using

organic paper of the Millipore variety. This type of filter material can conveniently be dissolved in strong acid to permit radiochemical analysis.

In some experiments iodine was also adsorbed on charcoal which was later heated to remove rare gases. Iodine was retained on the charcoal even at elevated temperatures, after once having been adsorbed. More recently, a new system containing charcoal maintained at 200°C was found sufficiently retentive for iodine adsorption and yet showed no retention of the rare gases, which are adsorbed separately at low temperatures.

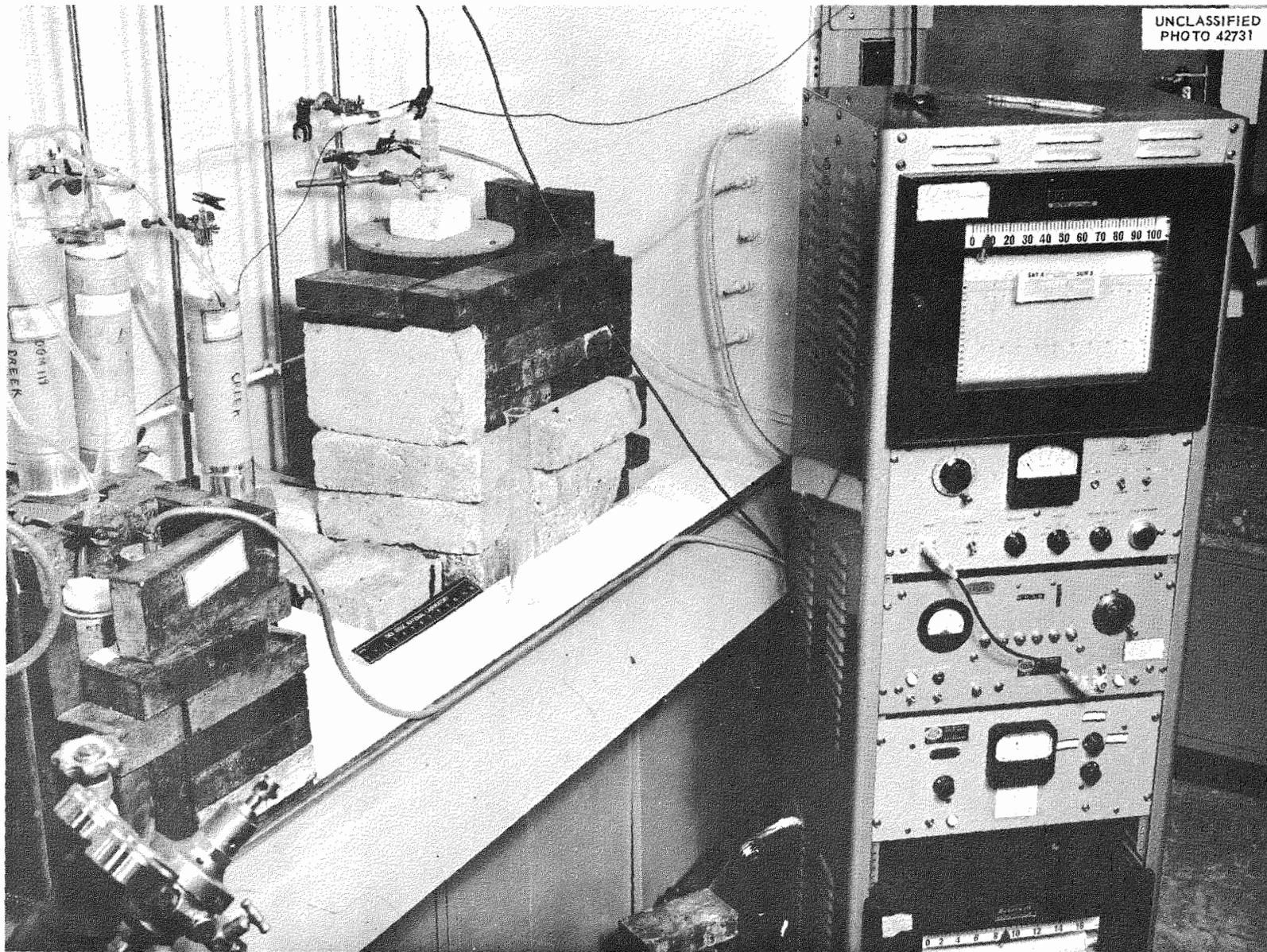


Fig. 3. Platinum Element Furnace for Long Term Melting.

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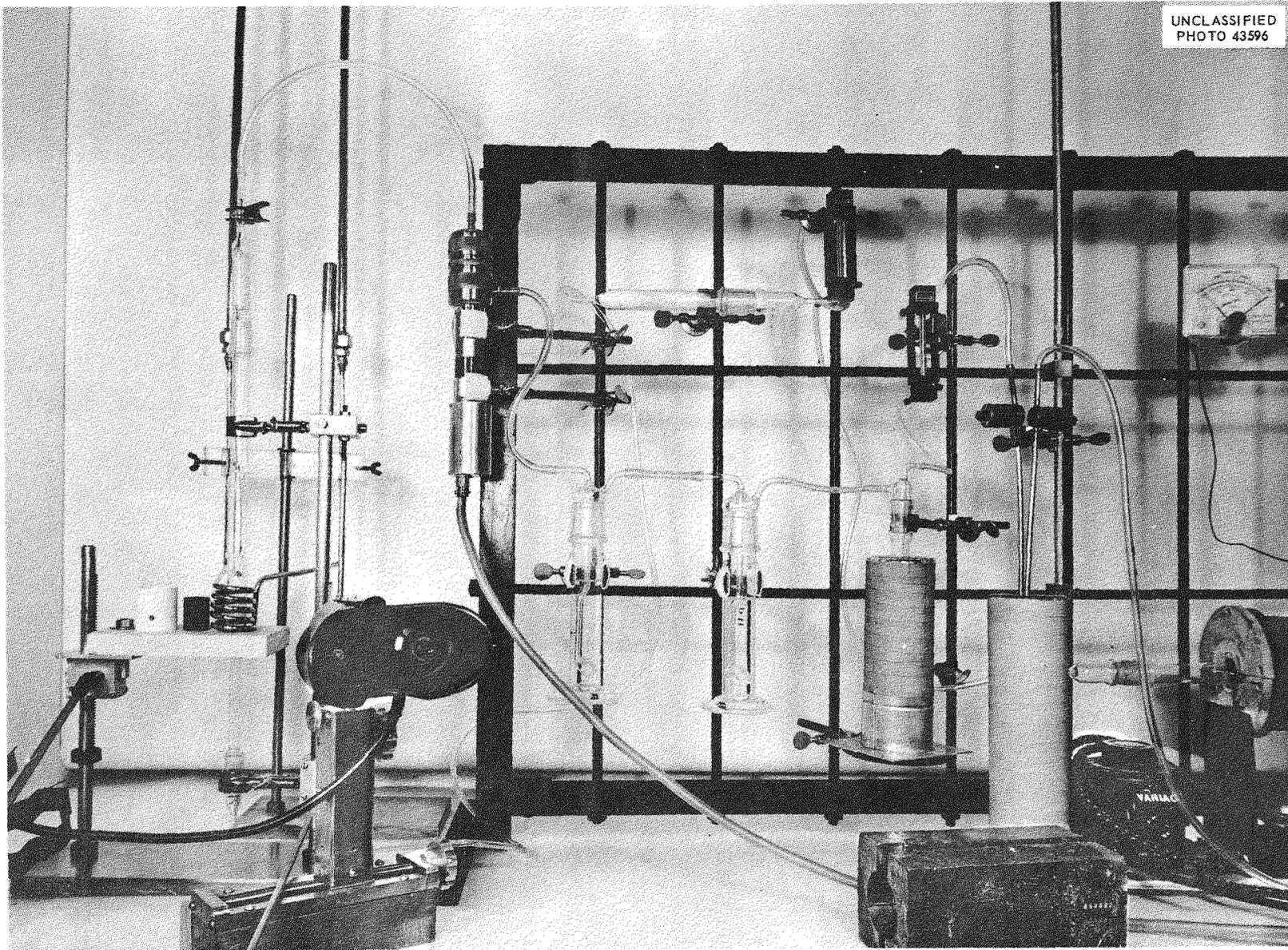


Fig. 4. Closeup of Apparatus for Inductive Melting of Reactor Fuel.

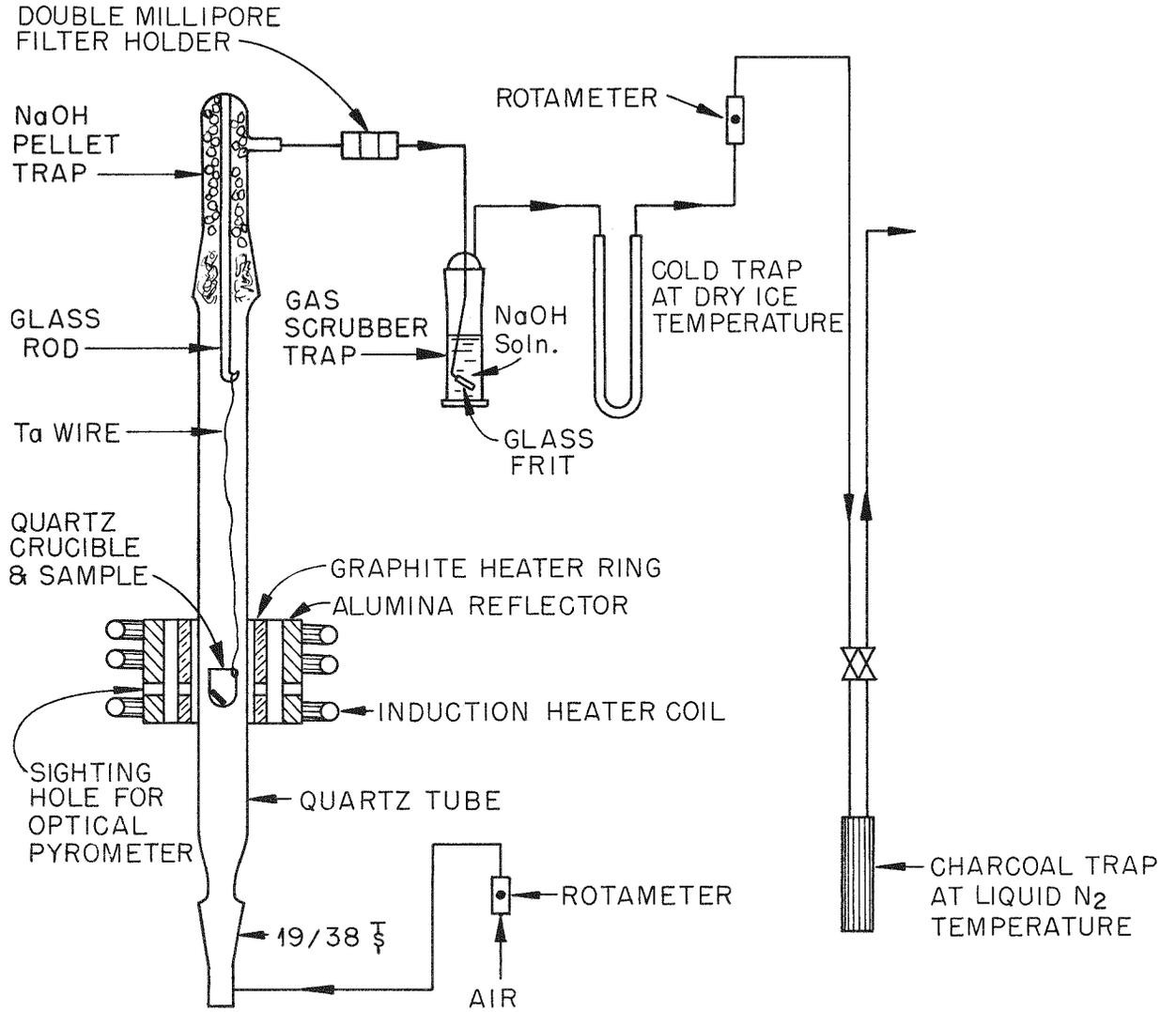


Fig. 5. Furnace and Traps for Induction Melting.

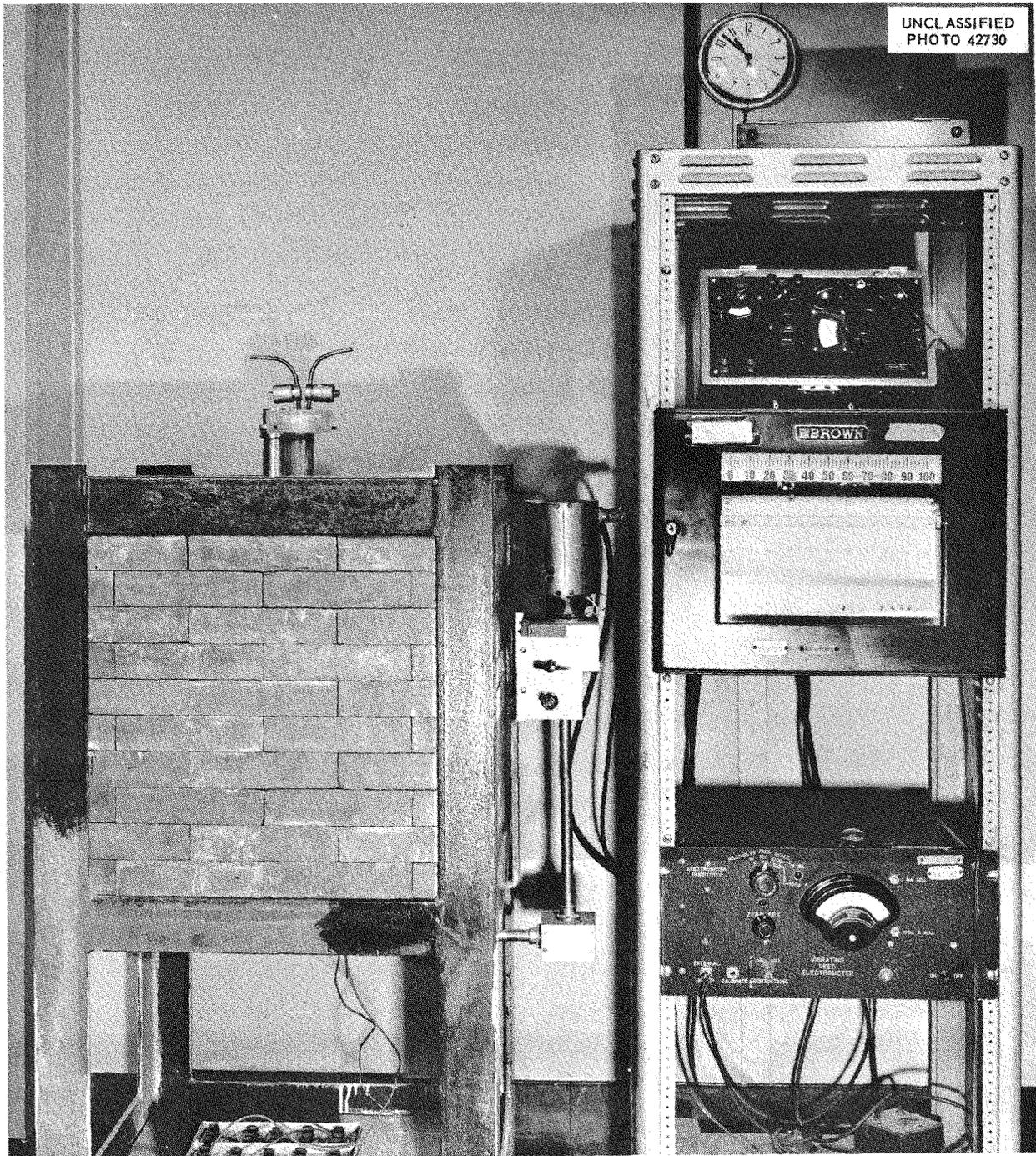


Fig. 6. Gamma Ionization Chamber and Electrometer.

**FISSION PRODUCT RELEASE FROM MTR-TYPE
ALUMINUM ALLOY**

The melting experiments with MTR-type aluminum alloy have all been performed using punchings from fully enriched plate. Because of its surface oxidation in air, the metal rarely gives a visible indication of having reached the molten state. As in the case of zirconium, the melt is confined in a semirigid framework of the oxide. The melting of the aluminum alloy is of interest in that relatively small amounts of the fission products are released at the fairly low melting temperature (700°C). The percentage is increased by elevating the temperature to 800°C, and by using a gas purging process the rare gases may be displaced quantitatively from the melt. On the other hand, the undisturbed melt shows a marked "squeeze effect" upon cooling which accounts for practically all of the gas actually released during melting. An illustration of this effect, which is probably related to a general outgassing process as the metal freezes, is shown in Fig. 7.

Tables 2 and 3 give a summary of the loss of iodine and the rare gases at different temperatures and for different periods of remaining molten. The data are all based on short irradiations and therefore on trace concentration of fission products. Most of the samples have been melted in air. It remains to continue the aluminum alloy program to other atmospheric environments and higher burnups.

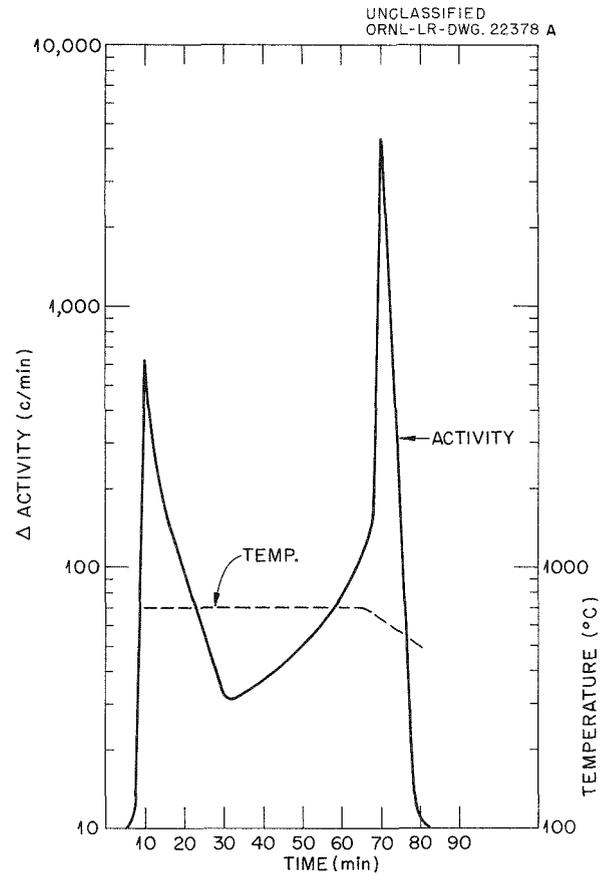


Fig. 7. Rate of Rare Gas Release from Al-U Pellet at 700°C, Showing Activity Peak on Cooling.

Table 2. Fission Product Volatilization from MTR Disks*

Run No.	Time In-Pile (hr)	Time Out-Of-Pile (hr)	Temperature of Run (°C)	Per Cent of Total Iodine Released	Per Cent of Total Released
1	13.5	7.25	700	1.2	0.45
2	13.5		700	0.88	0.41
3	60.5	6.33	700	0.27	0.30
4	16	2	700	0.49	0.15
5	60.5	5.5	700	1.8	0.82
				Av	0.93
6	13.5	7.7	800	5.3	2.26
7	13.5	9.25	800	4.9	2.06
8	16	5.25	800	4.18	1.10
				Av	4.79

*MTR pellets melted in air and held at 700 or 800°C for 1 hr.

Table 3. Release of Rare Gases by Rapid Melting of MTR Disks*

Run No.	Atmosphere During Melting	Per Cent of Rare Gas Released
1	Air	9.4
2	Air	8.4
3	Air	11.2

*Two minutes at 700°C.

Miscellaneous Experiments on Aluminum Fuel Plates

A question of some interest was whether a significant "dilution effect" could be observed upon the addition of inert metal to a fuel sample so that the total sample is melted at one time. At least a partial answer is that as far as the rare gases are concerned, the "dilution effect" is negligible, as is shown in Table 4. A comparison of Geneva-type fuel plate (UO₂ dispersion in aluminum) with MTR-type alloy, at least initially, shows a considerably better retention by the Geneva oxide plate. This is shown in the same table. These data will be extended later to several degrees of burnup in a new ORNL aluminum-U₃O₈ dispersion-type fuel plate.

FISSION PRODUCT VOLATILIZATION FROM APPR FUEL - A UO₂ DISPERSION IN STAINLESS STEEL

One of the first power producing reactors to be built near a populated center was the APPR

located at Fort Belvoir, Virginia. The stainless-steel-clad UO₂ dispersed fuel system was naturally among the first to require an early appraisal for fission product release. To a considerable extent, this type of fuel has been investigated in greatest detail.

For carrying out these extensive experiments, completely clad fuel miniatures (Fig. 1) were specially fabricated to be representative of actual reactor plate. Since time did not permit completing irradiations of fuel plate miniatures, samples with high burnup were punched from previously irradiated plate. The following experiments were conducted with one or both types of samples.

Fission Product Volatilization from APPR Clad Coupons Melted in Air or Steam

The melting of completely clad APPR coupons gave not too surprising results when compared with those from the lower-melting aluminum. The data presented in Table 5 result from relatively slow melting (1 to 2 min), however, without accurate time correlation with individual specimens. All samples required dissolution of the melt residue in aqua regia and collection of rare gases and iodine during the operation in an absorption train. All results are given by radiochemical analyses except the rare gas activity, which is an ionization chamber measurement.

Fission Product Volatilization from APPR Fuel After 25% Burnup

Results on melting APPR fuel punched from a previously irradiated test fuel assembly plate

Table 4. Release of Rare Gases from Geneva Oxide Fuel and MTR Aluminum Alloy in Air and Helium

Type of Plate* Melted	Number of Samples	Atmosphere	Fission Product Released (Av %)	
			Xe-Kr	Iodine
MTR	6	Air	13.3	
MTR**	7	Air	14.8	
MTR	10	He	15.1	
MTR**	14	He	20.8	
Geneva	3	Air	5.6	3 × 10 ⁻³

*Short irradiations of punched samples, trace concentration of fission products heated 5 min at 850°C.

**Sample sealed in aluminum tubing before irradiation.

suggest a significant burnup effect on the release of some fission products. At this time it is not clear whether the exposed edge of the fuel plate punchings accounts in any way for the significant increase in the amount of cesium vaporized. The relatively high "gross gamma" is largely a measure of cesium activity since the fuel plate had "cooled" for more than two years. These values will be compared in future work with similar results obtained from coupons. Here the steam atmosphere seems to depress slightly the release of volatiles, while helium causes increased release of both cesium and strontium. The data are given in Table 6.

Variations in the Amounts of Volatile Fission Products Released with Different Preheat Times for Melting

This experiment was designed to aid in explaining the rather wide scatter in data, particularly the variation in the amount of iodine released in previous duplicate melting experiments. It has been surmised that inherent in these experiments were possibilities for a considerable difference in total time-at-temperature from one run to another.

In order to correlate such time-to-melt with the release of activity, the following method was

Table 5. Fission Product Volatilization from APPR Clad Coupons* Melted in Air or Steam

Run No.	Atmosphere	Preheat Time To Melt (sec)	Rare Gases (%)	Gross Gamma (%)	Iodine (%)	Cs (%)	Sr (%)
1	Air	125	45	5.37	49		
2	Air	55	59	2.2	25		
3	Air	42	38.4				
4	Air	94	47.6	4	15.4		
5	Air	137	40	1.5	34		
6	Air	90	53.6	8	31	11	0.001
7	Air	144	61	6	41	13	0.001
8	Air	75	44				
9	Air	151	49.7	4.5	38		0.0001
10	Air	75	45.7	4.4	35	3.4	0.1
			Av 48.4	4.5	33.5	9.1	0.025
11	Steam		38.5	2.3	11.2	0.3	0.4

*Coupons irradiated in graphite reactor for one week.

Table 6. Fission Product Volatilization by Melting APPR Punched Disks, 25% Burnup

Run No.	Atmosphere During Melting	Maximum Temperature (°C)	Time To Melt (sec)	Per Cent of Total Activity Released				
				Gross Gamma	Iodine	Cs	Sr	Rare Gases*
1	Air	1575	13	0.4	-	12.0		-
2	Air	1650	15	0.2	-	5.8		-
3	Air	1575	47	17.3	-	68.3		-
4	Steam		45	16.8	-	74.9	0.06	-
5	He	1650	37	26.1	-	99.9	6.8	-

*Decayed through long cooling period, not reirradiated.

applied: The samples used were punched from a fully enriched APPR fuel plate. Some of the samples were reirradiated in the reactor for one week and allowed to "cool" for one week; from these the iodine and rare gas values were obtained. A second series of samples was melted without reirradiation for the cesium results. Different preheat times were obtained by using graphite heater rings of different thicknesses. The rate of heating by induction in the graphite ring varied inversely as the graphite thickness. The "melting time" was taken as the time required for heating the sample from room temperature to its melting point. Accurate melting times were not obtained because of some difficulty in electric coupling with the different heaters. Approximate times are given.

The results are still observed to scatter; however, the trends established are definitely related to the heating period. The data for iodine and rare gases are given in Table 7 and Fig. 8. The summary data for cesium are given in Table 8 and Fig. 9.

The preheat effect is so marked in the case of the cesium that some thought has been directed toward an explanation. The effect of the exposed edge, which must be highly enhanced in the UO_2 dispersion-type plate, has not been ruled out. In fact, it has been observed in some incomplete work that the fission gases are completely released from similar high-burnup APPR punchings at temperatures still below the melting point.

A possible contributor to the preheat effect is a chemical reaction between UO_2 and some of the constituents added to APPR fuel, for example, silicon or boron, which may give a thermite-type

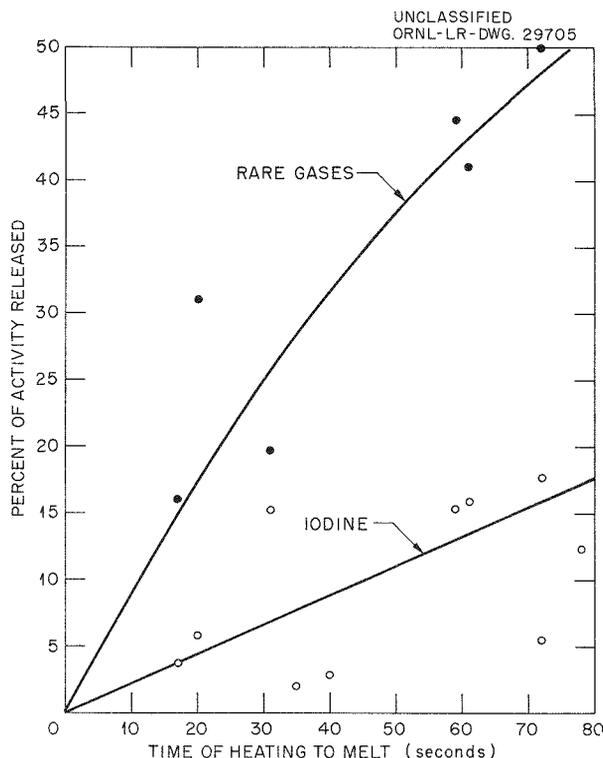


Fig. 8. Release of Fission Products as a Function of Heating Rate.

Table 7. Variation in the Amounts of Iodine and Rare Gases Released from APPR Disks* with Different Preheat Times

Sample No.	Thickness of Heater Ring (in.)	Preheat Time (sec)	Pyrometer Reading at mp ($^{\circ}C$)	Per Cent of Total Rare Gases Released	Per Cent of Total Iodine Released
1	0.0625	17	1300	16	3.7
2	0.125	20	1565	31.3	5.7
3	0.25	31	1500	19.4	15.2
4	0.25	59	1618	44.6	15.3
5	0.312	61	1550	41.0	15.9
6	0.312	72	1521	50.7	17.6
		Av 43.3		34.8	12.2

*20% burnup.

reaction such as was observed during the manufacture of the initial Geneva aluminum fuel. This theory is being explored by x-ray examination of melted stainless steel-UO₂ for evidence of new intermetallic compounds.

Table 8. Variation in the Amount of Cesium Released from APPR Disks* with Different Preheat Times

Sample No.	Thickness of Heater (in.)	Preheat Time (sec)	Per Cent of Total Cesium Released
1	0.062	6	15.7
2	0.125	11	18.7
3	0.25	20	34.8
4	0.31	34	64.0
5	0.31	42	72.2
		Av 22.6	41.0

*30% burnup.

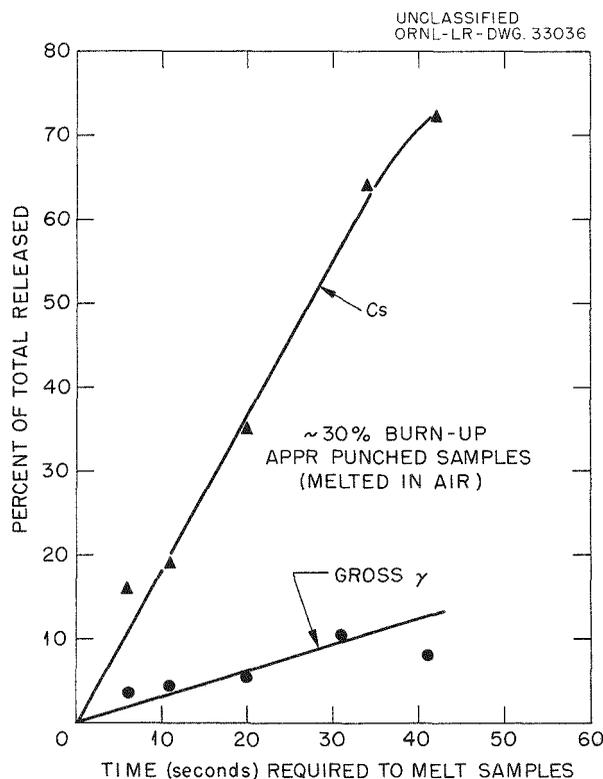


Fig. 9. Correlation of Percentage Release of Cesium with Total Heating Time.

Diffusion of Fission Products Through APPR Cladding at Temperatures Below the Melting Point

Because of its significance in reactor design, diffusion through the cladding has been postulated as a possible mechanism for accumulating significant amounts of radioactivity at temperatures considerably below the melting point. In order to investigate the magnitude of this effect, APPR clad coupons have been heated in a nitrogen gas stream at 900°C for periods of 7, 22, and 32 days. In all of these experiments, no measurable activity was released. Details are given in another report.¹⁷

By raising the temperature to about 1400°C, a slight release (~0.01%) of the fission gases (Fig. 10) was observed in helium after several hours. Upon examination, the sample appeared to have undergone some chemical change in the fuel-bearing area, since this portion was clearly

¹⁷L. P. Crocker and G. E. Creek, *Fission Product Diffusion Through Clad Cermet Fuel Elements*, ORNL CF-57-12-69 (Dec. 16, 1957).

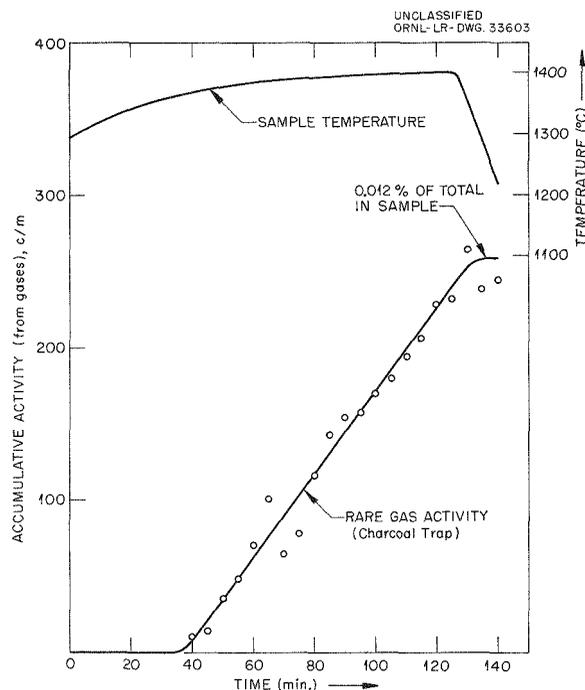


Fig. 10. Release of Rare Gas Activity from APPR Coupon at Temperatures Below the Melting Point (Heated in Helium).

outlined after the heating period. Several experimenters have measured diffusion rates of fission gases through uranium or uranium oxide with out-of-pile heating. Recently, published diffusion rates measured under in-pile conditions have indicated a highly accelerated process in UO_2 . This same effect may be observed later in metal-clad systems and warrants some attention.

Suppression of Fission Product Release from APPR Fuel by Means of Ceramic Coating

One aspect of the release of fission products from fuel plate seems to bear a relation to the amount of oxidation. It seems logical that in the extreme case in which the base metal is completely oxidized, the volatile fission products would be released as a result of the increase in porosity and particle subdivision, although they could otherwise be retained because of their slow rate of diffusion. A simple experiment was designed to demonstrate such an effect. Fuel coupons were coated with a thin (5-mil) layer of a high-temperature ceramic (NBS-A418) which upon melting would serve very much in the same way as a welding flux to prevent oxidation. Figure 11 is illustrative of the protection from oxidation afforded in this way. In the photograph the clean, bright metal of the coated sample melted in air shows that no oxidation occurred.

After irradiation of the coupons (to only trace concentration of fission products), both plain and coated samples were melted. The results, in Table 9, show no striking improvement for the coated samples; however, since the preheat times were short, the release values are all low. Any beneficial effect was apparently lost in the poor statistics.

RELEASE OF FISSION PRODUCTS FROM HTRE-DIRECT-CYCLE FUEL PLATE

With the use of approximately the same equipment that was described for inductive melting of APPR fuel, small clad specimens of HTRE fuel plate were melted in air following a short irradiation. The results based on two samples melted in air with about 30 sec of preheat are given in Table 10. A similar experiment but with a long molten period of 4 hr showed the expected increase, accounting for almost quantitative release of iodine and the gases. The cesium value was

lower than expected, perhaps because of the low burnup. These data are shown in Table 11.

RELEASE OF FISSION PRODUCTS ON HEATING UO_2 BELOW THE MELTING POINT

The work of Lustman¹⁸ and others¹⁹ has established to a considerable extent the behavior of the rare gases on irradiation of UO_2 in terms of diffusion coefficients. Recent studies by Barney and Wemple²⁰ have demonstrated extensive plastic flow in UO_2 at high burnup. These same studies suggest possible escape routes for fission products through extensive cracking of cooler outer areas surrounding molten center sections.

At present, anticipated use of UO_2 has displaced practically all other fuels as an economical civilian power reactor fuel. Systems are presently designed or built for encapsulation of UO_2 in rod, ring, and plate form in both stainless steel and zirconium. The design of fission product release studies for UO_2 in these systems has progressed only as far as melting off the cladding and to the extent of heating unclad, slightly irradiated UO_2 for sustained periods all at temperatures considerably below the melting point ($\sim 2750^\circ C$). The results of this amount of examination are all pleasing in that little of the fission gases and practically nothing else was vaporized in the process. These values are reported tentatively in Table 12.

Ignition of UO_2 by Melting Zircaloy Cladding

One possible type of miniature Zircaloy-clad UO_2 rod specimen (PWR) is the small capsule shown in Fig. 1. One such experimental sample was prepared and subjected to induction heating to $2000^\circ C$, which was sufficient for decladding but considerably below the melting temperature of UO_2 . On cooling, the metal exhibited considerable cracking and sparking, and on examination it was found that the UO_2 had been ignited. A fairly significant portion of the UO_2 burned visibly after

¹⁸B. Lustman, *Release of Fission Gases from UO_2* , WAPD-173 (March 1957).

¹⁹J. M. Markowitz, R. C. Koch, and J. A. Roll, *Release of Fission Gases from Irradiated Uranium Dioxide*, WAPD-180 (July 1957).

²⁰W. K. Barney and B. D. Wemple, *Metallography of Irradiated UO_2 -Containing Fuel Elements*, KAPL-1836 (Jan. 15, 1958).

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CERAMIC COATED
TYPE 304 STAINLESS STEEL
MELTED IN HELIUM

CERAMIC COATED
TYPE 304 STAINLESS STEEL
MELTED IN AIR

TYPE 304 STAINLESS
MELTED IN AIR

Fig. 11. Ceramic Coating Used To Control Oxidation for Melting Stainless-Steel-Clad Fuels.

650
650

Table 9. Comparison of Ceramic Coated and Plain APPR Coupons for Release of Fission Products

Experiment No.	Type of Coupons*	Time of Melting (sec)	Apparent Temperature (°C)	Gases Released (%)	Iodine Released (%)	Gross Gamma (%)	Cs (%)
1	Plain	21	1650	9	0.4	0.7	10.8
2	Coated	20	1575	15.9	1.8	0.4	1.8
3	Plain	20	1635	25	3.4	1.6	0.5
4	Coated	27	1610	16.7	1.3	0.6	0.4

*Irradiated to trace concentration of fission products.

Table 10. Release of Fission Products from Quick Melting of HTRE GE Fuel Specimens

Experiment No.	Time Required To Melt Sample* (sec)	Per Cent of Total Rare Gases Released	Per Cent of Total Iodine Released	Per Cent of Total Cesium Released	Per Cent of Total Strontium Released
1	30	9.7	4.18	0.7	
2	32	10.2	4.42	0.3	1.1×10^{-4}

*Melted with induction heater, using graphite susceptor ring, in air. Irradiated to trace concentration of fission products.

Table 11. Release of Fission Products from HTRE Fuel* After 4 hr Above the Melting Temperature

	Gross Gamma (%)	Iodine Gamma (%)	Cs Gamma (%)	Sr Beta (%)
Basic wash	3.7	37.4	0.3	0.004
Acid wash	2.8	18.7	0.6	0.006
Fluoride wash	0.3	0.2	0.6	0.004
Pellet trap	1.25	42.8	0.06	0.0004
Scrubber trap	0.001	0.005		
Total release**	8.06	99.06	1.66	0.01

*Irradiated to trace concentration of fission products.

**A comparison of the charcoal gas trap used in the melting experiment with one used in the dissolving of the second sample indicates a loss on melting of 76.9% of the xenons and kryptons.

Table 12. Release of Fission Gases on Heating Sintered UO_2 at Temperatures Below the Melting Point

Experiment No.	Time of Heating (min)	Maximum Temperature ($^{\circ}C$)	Per Cent of Total Rare Gases** Liberated	Method of Heating
1	330	1300	0.019	Pt resistance furnace
2	0.5	1638	1.27	Induction
3*	0.1	1600	1.0	Induction
4*	0.1	1600	0.4	Induction

*Encased in stainless steel capsules; stainless steel melted in air.

**Trace concentration of fission products.

cooling. Plans are being made to melt UO_2 samples by means of a special high-temperature furnace.²¹ The effect of ignition of UO_2 , if found recurrent, will strongly influence fission product release.

RELEASE OF FISSION PRODUCTS BY MELTING STR-ZIRCALOY PLATE, 15% BURNUP

Experience with melting Zircaloy plate of the STR type has been limited to the use of punched samples from a large experimental plate. The melting experiments have been performed in two ways (see Fig. 12). In the first experiment, punched samples were melted by the radiant graphite susceptor in the induction furnace. The release values, which are considered subject to criticism because of experimental difficulties, are summarized in Table 13.

In the second, more recent series, the samples were encapsulated to avoid as much as possible the exposed edge problem and to afford sufficient mass for direct coupling. The capsule and sample weighed about 5 g. Melting was accomplished in 30 sec in the equipment shown schematically in Fig. 13. After washing out the equipment and dissolving the residue in an $HF-HNO_3$ mixture, radiochemical analyses were obtained on all fractions. These results of melting in air and in steam are given in Table 14. Some significant effect on release ratios is apparent in the variations observed in air, steam, and helium. The irregularity in the values obtained for cesium

seems to have been suppressed by the encapsulation. The correlation of these data with those being obtained by MSA on a somewhat larger scale is not seriously outside experimental limits. At present, our results obtained for strontium are somewhat less than those found by MSA.² Other values seem to be in agreement.

It was of interest to examine the various volatile fractions for apparent gamma activity by means of a gamma-ray spectrometer. In doing this, any major contributor can be readily located. In Figs. 14, 15, and 16 all of the principal gamma activities are identified in the separate fractions according to energy.

It is interesting to note that the cesium release is rather strongly suppressed in the higher melting Zircaloy as compared with the stainless steel- UO_2 dispersion, suggesting that the alloy fuel is less susceptible to radiation damage at high burnup.

A strict comparison of the data in the two tables cannot be justified since experimental difficulties taxed the facilities in use at the time the data in Table 13 were taken. A great deal more confidence is felt for the results in Table 14. It is expected that in continuing the work more detailed studies of the effects of the atmosphere and the preheat time will be carried out.

PARTICLE-FORMING FISSION ELEMENTS RELEASED FROM APPR FUEL PLATE MELTED IN AIR

The most recent work has been directed toward an examination of the meltoff gas or vapor for particle-forming fission products in addition to the volatile components already characterized.

²¹M. R. Null and W. W. Lozier, *Rev. Sci. Instr.* **29**, 163-70 (1958).

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S T R FUEL PLATE
PARTIALLY MELTED



ZIRCALLOY CAPSULE
&
PUNCHED FUEL SAMPLE



ZIRCALLOY CAPSULE
AFTER MELTING

Fig. 12. Zircaloy Fuel Samples for Melting Experiments.

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Table 13. Fission Product Volatilization by Melting STR-Zircaloy Punched Disks, * 15% Burnup

Run No.	Atmosphere During Melting	Maximum Temperature (°C)	Time To Melt (sec)	Per Cent of Total Activity Released				
				Gross Gamma	I ₂	Cs	Sr	Rare Gases
1	He	1900	33	11.6	40.9	26.3		95.6
2	He	2025	37	11.2	38.8		0.1	99.4
3	He			8.0	80.8	65.9	0.4	95.7
4	Air	1925	52	4.5	53.2	4.5	0.2	95.7
5	Steam	1925		1.3	53.5	0.6	0.04	98.0

*Exposed edge samples, melted by use of graphite susceptor ring.

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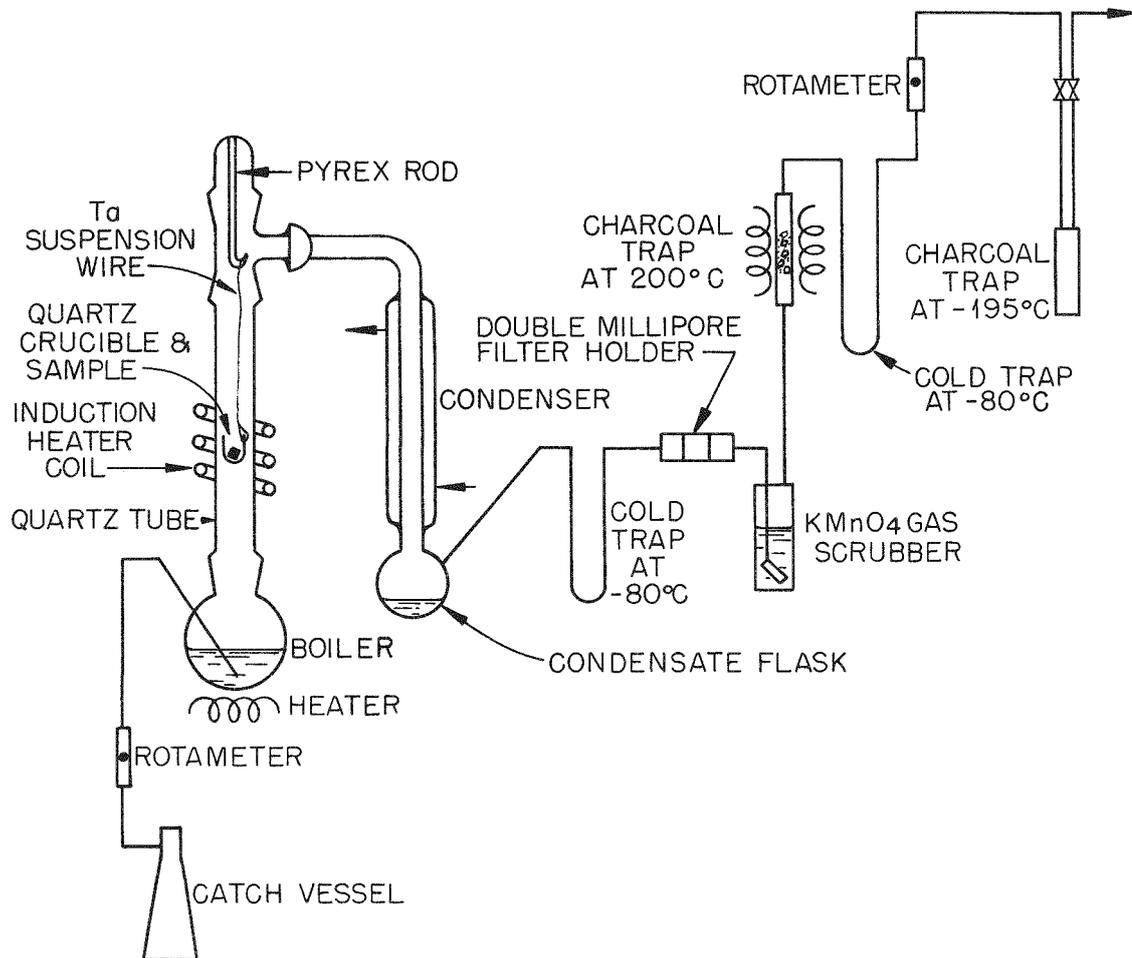


Fig. 13. Apparatus for Melting Fuel Capsules in Steam.

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Table 14. Fission Product Volatilization by Melting Encapsulated Zircaloy Punched Disks, 15% Burnup

Run No.	Atmosphere During Melting	Heating Time ^a (sec)	Maximum Temperature ^b (°C)	Per Cent of Total Activity Released						
				Rare Gases	Gross Gamma	I ₂	Cs	Sr	Ba	Ce
10-10	Air	12.5	1705	100	2	28.0	10.0			
10-15	Air	16		100	2.2	32.4	11.3			
10-16 ^c	Air	12	1750	100	6.4		11.7			
11-25	Air	30	1705	100	2.5	14.4	7.3	0.9	0.11	0.0047
11-26	Air	30	1800	100	4.1	30.0	12.9	0.8	0.3	0.004
		Av 20.1	1740	100	3.4	26.2	10.6	0.85	0.2	0.0043
10-20	Steam	33		100	5.3	12.8	8.9	2.4	0.25	0.05
10-22	Steam	32	1775	100	5.5	52.4	23.0	3.1	1.2	0.01
11-11	Steam	18		100	7.8	66.4	21.8	4.8		
11-12	Steam	31	1750	100	7.2	55.8	24.4	1.3		
11-14	Steam	35	1750	100	4.8	57.4	12.9	0.2		
11-21	Steam	30.5	1750	100	8.0	45.2	19.4	0.2	0.049	
11-24	Steam	31.5	1730	100	5.7	36.3	20.3	0.8	0.20	
		Av 30	1750	100	6.3	46.6	18.7	1.8	0.4	0.03

^aOptical pyrometer temperature.

^bPunched disk not reirradiated.

^cSample usually melted in approximately 12 sec.

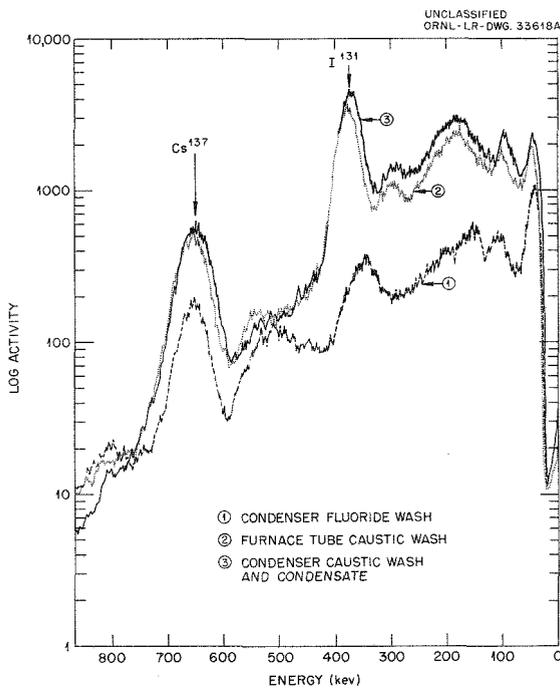


Fig. 14. Gamma Scan of Activity Released when Zr-U Alloy Is Melted in a Steam Atmosphere.

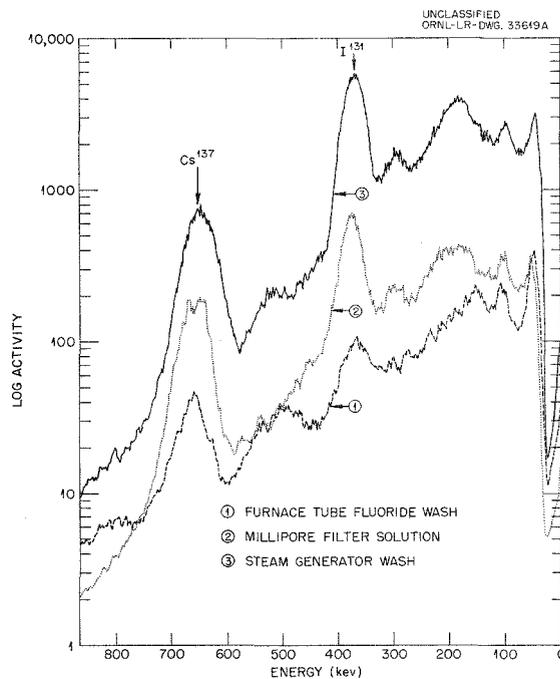


Fig. 15. Gamma Scan of Activity Released when Zr-U Alloy Is Melted in a Steam Atmosphere.

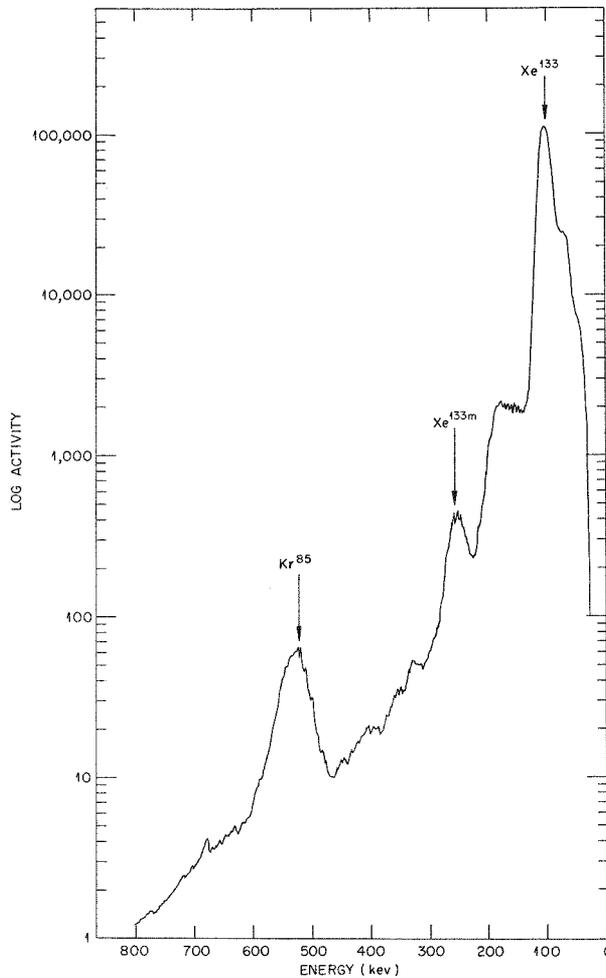


Fig. 16. Gamma Scan of Rare-Gas Activities from Melting Zr-U Fuel in Steam Atmosphere (Charcoal Trap).

Table 15 illustrates the relative probability of finding the most important fission element activities in a radioactive cloud from a meltdown. In this experiment, $\frac{3}{8}$ -in. disks were punched from APPR stainless steel- UO_2 plate after having a 20% burnup irradiation and having two years cooling time.

The values observed for all elements except cesium are so low that they are not clearly different from zero; therefore, it was not surprising when a similar result was found in the following experiment. Using the induction-type heating method and an APPR fuel plate containing only long-lived fission products as the source, samples were melted in an air stream which carried the air-borne materials to a series of filters of Millipore and CWS papers. The porosity of the

Table 15. Particle-Forming Fission Elements Released by Melting* APPR Fuel Plate

Fission Product	Radiation	Average Percentage Released
Cs^{137}	Gamma	11.0
Zr^{95}	Gamma	0.05
Ce^{144}	Beta	0.008
$\text{Sr}^{89,90}$	Beta	0.006

*Preheat time 27 sec, maximum temperature 1525°C.

paper selected was such that an efficiency greater than 99% was expected for particles down to 0.05 μ .

In all cases the activity collected was found to be pure Cs^{137} , and this was essentially completely stopped on the 5- μ (pore diameter) paper. Table 16 summarizes the results of the collection of cesium on a two-stage filter.

It is known that cesium oxide is thermodynamically unstable at the temperature of molten stainless steel, and therefore it is presumed that it vaporizes as elemental cesium only to reform as the oxide at a lower temperature. The deliquescent character of the cesium hydroxide confuses efforts to measure particle size.

Filter Collection of Iodine Released from Melted APPR Fuel

The released iodine has been characterized in terms of oxidation states represented by finding a decidedly higher efficiency for removal in a sodium bisulfite scrubber over a sodium hydroxide scrubber. From this it is inferred that the majority is vaporized as elemental iodine. The fact that some iodine passed through both scrubbers suggests that while the major portion is present as elemental iodine instead of reduced iodine, some fraction may also exist in a higher oxidation state.

It was of interest then to attempt to collect the iodine on filters as was done with cesium, since these two elements present the predominant portion of the biological hazard created. Using both mineral (CWS) and organic filters,²² air was drawn

²²Millipore Filter Corp., Watertown, Mass.

from the induction melting of a short-period-irradiated APPR fuel disk through two successive filters, an NaOH pellet absorber, and a cold charcoal trap. The fact that iodine was found distributed throughout the entire system was expected, but that a large percentage was collected on the first filter was surprising. The results are shown in Table 17.

CONCLUSIONS

1. The composition of the fission products released to the atmosphere from a quiet meltdown of reactor fuel is limited mainly to the "volatile constituents" whose boiling points are lower than the melting temperature of the fuel.

2. The percentages released are relatively low for "threshold melting" and are caused to increase mainly by increasing the length of "preheat time," the concentration of fission products, and degree of oxidation. Contributing factors are the nature of the surrounding atmosphere, exposure of the

fuel-bearing edge of the sample, size of the sample, and type of fuel.

3. The most important result seems to be a rather close correlation of fission product release with fuel melting temperature (with the exception of cesium), especially when melted in air. This correlation, which may be fortuitous, is shown in Fig. 17. The significance of time-of-preheat in stainless steel-UO₂ before melting is much more than was expected and is not presently understood. Since this value corresponds to the loss-of-coolant afterglow period, it should be given consideration in hazards evaluation.

4. No significant effect has been observed to indicate a gross metal-water reaction when zirconium alloy, aluminum alloy, or stainless steel-UO₂ is melted in steam. The important fission products released are the rare gases, iodine, and cesium (and under special conditions, strontium), in this order. The significance of these in terms of percentages of the gross beta activity is given in Table 18.

Table 16. Filtration of Particulate Cesium from Melted APPR Disks*

Run No.	First Filter	Second Filter	Ratio of Activity Collected
			$\frac{\text{Cs on first}}{\text{Cs on second}}$
1	0.8 μ (Millipore)	0.8 μ (Millipore)	5×10^5
2	5 μ (Millipore)	0.8 μ (Millipore)	6×10^4
3	0.3 mm (HV-70**)	0.8 μ (Millipore)	340

*15% burnup.

**Mineral fiber, paper, 0.3 mm thickness.

Table 17. Filtration of Released Iodine* from Melted APPR Disks

Run A		Run B	
Filter or Adsorbent in Series	Per Cent Collected	Filter or Adsorbent in Series	Per Cent Collected
5- μ paper (Millipore)**	91.7	1-mm paper (CWS-6)	70.6
0.8- μ paper (Millipore)	0.6	0.8- μ paper (Millipore)	2.1
NaOH (pellets)	2.5	NaOH	11.4
Charcoal (-1950°C)	5.2	Charcoal	15.8

*Trace concentration.

**5 μ mean pore diameter, 150 μ thick.

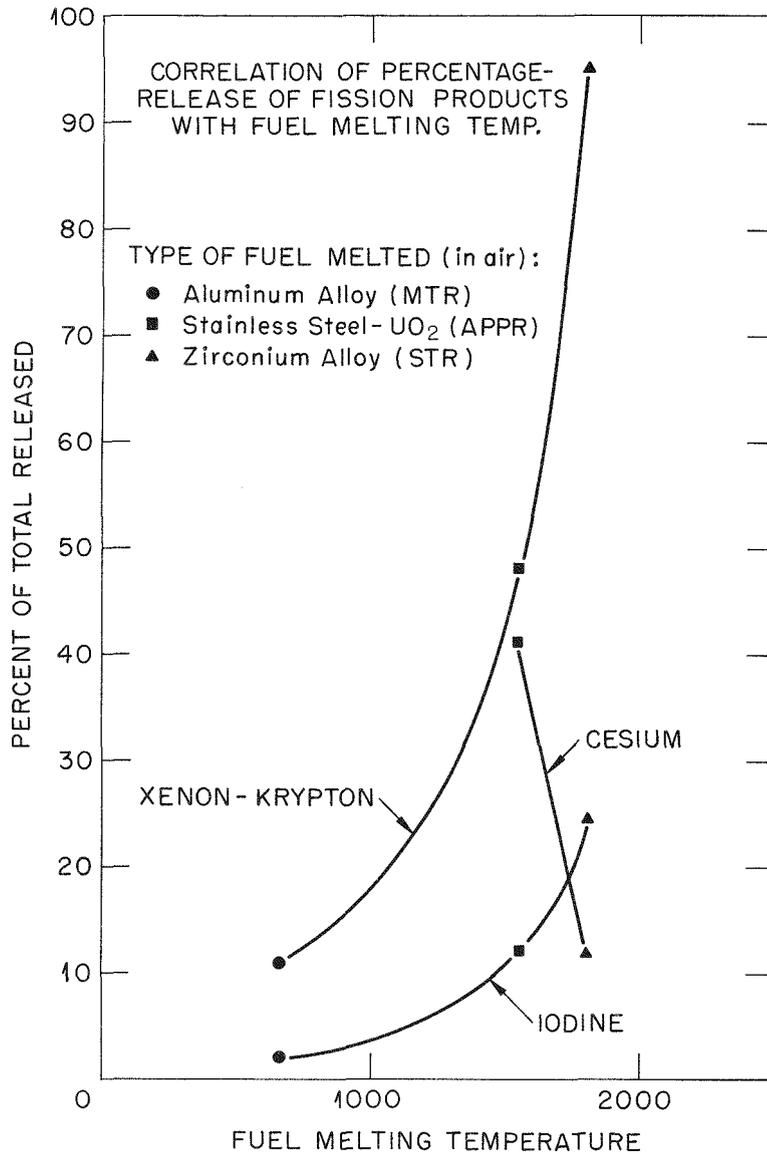


Fig. 17. Correlation of Percentage Release of Fission Products with Fuel Melting Temperature.

Table 18. Calculated Beta Activity of the Fission Products^a
Per cent of total by element^b

		Cooling Time			
		10 min	10 hr	10 days	100 days
5 Days Irradiation					
Group I-A	Xe	6.0	11.0	8.0	0.2
	Kr-Rb ^c	3.4	0.8	0.02	0.06
	I-Br	9.0	15.0	8.5	0.1
	Cs ^c	3.5	0.001	0.03	0.09
Group I-B	Ba	5.0	2.5	10.0	0.15
	Sr	6.0	4.5	3.3	11.0
Group II	Te-Se	7.0	4.0	4.8	0.05
	Mo	14.0	6.2	4.8	0.00
	Tc	0.3	0.6	0.5	0.02
Group III-A	Y	11.0	10.0	3.3	14.0
	La	11.5	3.3	12.0	0.9
	Ce	5.0	8.2	8.0	16.0
	Pr	3.6	3.8	11.0	8.0
	Nd	8.0	1.2	6.0	0.06
	Pm	0.6	1.6	1.2	9.0
Group III-B	Zr	5.8	13.0	7.0	15.0
	Nb	2.9	7.5	1.3	17.0
	Ru	0.6	0.8	3.0	5.5
	Rh	0.6	2.2	5.5	6.5
30 Days Irradiation					
Group I-A	Xe	6.0	10.0	4.5	0.1
	Kr-Rb ^c	3.0	0.7	0.02	0.008
	I-Br	10.0	13.0	5.7	0.04
	Cs ^c	3.0	0.001	0.04	0.24
Group I-B	Ba	5.8	5.0	10.5	0.3
	Sr	5.8	4.2	4.5	8.5
Group II	Te-Se	6.8	4.0	2.2	0.1
	Mo	16.0	5.6	2.5	0.0
	Tc	0.4	0.6	0.3	0.01
Group III-A	Y	10.0	8.0	5.5	14.0
	La	6.6	6.0	12.0	0.7
	Ce	5.7	8.2	10.2	14.0
	Pr	4.5	6.0	12.0	10.0
	Nd	1.4	2.5	4.3	0.02
	Pm	0.6	1.5	1.7	9.0
Group III-B	Zr	5.5	10.0	5.7	14.0
	Nb	2.7	5.3	2.6	23.0
	Ru	1.0	1.7	4.5	6.0
	Rh	1.3	3.3	5.5	6.0

^aMelting behavior:

- Group I-A High volatility; 10-100% released
- Group I-B Volatile in vacuum, nonvolatile in air; 1-10% released
- Group II Variable; alloying; low volatility
- Group III-A Low volatility; rare earths; refractory; 0.1% released
- Group III-B Low volatility; alloying and refractory; 0.1% released

^bFrom F. J. Keneshea, Jr., A. M. Saul, and C. T. Young, *The Distribution of Tracer Plutonium and Fission Products Between Molten Uranium and Solid Uranium Oxide, Carbide and Nitride*, NAA-SR-1032 (Sept. 15, 1954).

^cData extended.

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