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Abstract

Irradiated uranium aluminide fuel plates of 40 % U-235 enrichment were heated for the determination of fission products released under flowing helium gas at temperatures up to and higher than the melting point of fuel cladding material. The release of fission products from the fuel plate at temperature below 500 °C was found negligible. The first rapid release of fission products was observed with the occurrence of blistering at 561±1 °C on the plates. The next release at 585 °C might be caused by melting of the cladding material of 6061-Al alloy. The last release of fission product gases was occurred at the eutectic temperature of 640 °C of U-Al_x.

The released material was mostly xenon, but small amounts of iodine and cesium were observed.

Introduction

Studies on the release of fission products from a plate-type fuel are not abundant and most of the early data were concentrated on uranium metal alloy fuels[1,2] or UO_2.[3] Little data on U-Al alloy fuels,[4,5,6,7] have been reported. Especially scarce were data on U-Al_x dispersion fuels.[8] The detailed evaluation of the amounts of fission products released from irradiated fuel plates is required for the safety analysis of hypothetical accident conditions when the Kyoto University High Flux Reactor (KUHFR) will be operated using the fuel plates of 45 % enrichment uranium-235 in the future. For the sake of high power operation of the KUHFR at 30 MW, the development of the fuel plates with reduced U-235 enrichment and high content of uranium are required to keep

the fission density in fuel plates unchanged from the case of high enrichment uranium.

The main purpose of this experiment is to measure the threshold temperature at which a massive release of volatile fission products occurs and to estimate the release rate of traces of fission products at temperatures below the melting point of cladding material in as much detail as possible. The other objectives of this experiment are to examine the degree of mechanical deformation of the fuel plate after heating and to measure the quantities of fission products released at temperatures close to and higher than the melting point of fuel cladding material.

Experimental

Sample and Irradiation

The miniature fuel plates used for these experiments were fabricated by EG&G Idaho, Inc. and irradiated in the Oak Ridge Research Reactor (ORR) at Oak Ridge National Laboratory (ORNL). The plates were rectangular, 115 mm long and 50 mm wide. They consist of a layer of uranium aluminide (UAl_x) enriched to 40.24 % U-235 by weight, dispersed in aluminum and sandwiched between 6061-aluminum alloy claddings. The characteristics of the four plates used in the experiment are given in Table 1.

Table 1. Characteristics of the Samples

Run No.	Plate No.	Plate Weight (g)	Plate Thickness (mm)	Cladding Thickness Average (mm)	Cladding Thickness Minimum (mm)	^{235}U (g)	Uranium Density (Mg/m^3)
A	E-141	22.7557	1.295	0.38	0.27	1.901	2.217
B	E-101	23.7429	1.295	0.30	0.20	2.201	1.937
C	E-116	29.0746	1.549	0.34	0.21	3.173	2.306
D	E-114	28.2533	1.549	0.35	0.23	2.649	1.953

These plates were irradiated for an equivalent of 275 full-power days (fpd) from July 18, 1980 until July 10, 1981 in position E7 (217 fpd) and position C3 (58 fpd) of the ORR, a 30 MW water moderated reactor. This irradiation corresponds to about 60 % burnup of the initial U-235. Flux monitors indicated a thermal flux of 0.6×10^{14} to 0.8×10^{14} $n/cm^2 sec$ at the beginning of irradiation when the plates were located near the top of the core and 1.0×10^{14} to 1.3×10^{14} $n/cm^2 sec$ when the plates were moved 121 mm closer to the core midplane. These plates and their irradiation history are described in more detail by Senn and Martin.[9]

Equipment

The work of the release experiment of fission products was carried out in a manipulator hot cell in ORNL.

An electric furnace with nichrome heater was used to heat the miniature plate. The dimension of the heated zone was 178 mm (7 in.) in length and 76 mm (3 in.) in diameter. The furnace was shielded to reduce the radiation background at the radiation detector. Lead, 104 mm (4 in.) thick, was used to attenuate the direct radiation from the intensely radioactive fuel plate. The remainder of the furnace was shielded by 6.4 mm (1/4 in.) thick lead sheet.

A quartz tube, 64 mm (2.5 in.) I.D. and 356 mm (14 in.) long, was fitted inside the furnace. Two chromel-alumel thermocouples, sheathed in stainless steel, were inserted into the tube from the back end through a small 5 mm I.D. quartz tube. Helium gas was also introduced through the same small tube. The other front end of the tube was connected to the fission product collection system by a quartz ball joint.

The miniature fuel plate was placed on a slant bed in the quartz tube. The sheathed hot junction of a thermocouple was set in light pressure contact with the surface of fuel plate at its middle to measure the fuel plate temperature. The temperature was recorded by a single point recorder, calibrated by the ORNL Instrument Standard Section. The recorder could be read with an accuracy of 2°F (1.1°C). The other thermocouple was located above the plate and used to control the furnace temperature.

The experimental work was divided into two parts. The first part of the experiments was concerned with the measurement of a minute fission product release at fuel temperatures below the melting point of the aluminum cladding, while the second part with the melted fuel plate. The equipment for the first part was designed to trap and measure very small traces of Xe-133 and I-131. The equipment used in the second part of the work was setup to trap the relatively large quantities of Xe-133, I-131 and other volatile fission products that might be released at higher temperatures. A photograph of the actual setup of the equipment used in the first part of the experiments is shown in Photo. 1.

A sample trap used for the first part experiment consisted of glass column and activated coconut charcoal (20 g). A larger charcoal trap for backup was also used to collect any volatile fission products that might be passed through the sample trap.

The accumulation of fission products in the sample trap was detected by an ionization chamber attached to the cryostat containing the fission product trap. The ionization chamber and the cryostat were shielded by a 3.2 mm (1/8 in.) thick lead sheet.

Procedure

Helium gas was flowed over the plate and then through the sample trap, which was cooled to liquid nitrogen temperature. The flow could also be diverted to an alternate fission products trap by controlling the petcocks.

After the collection of fission products, the sample traps were removed and placed on a Ge(Li) detector to allow efficient counting of traces of Xe-133 or I-131 that might be released from the fuel plate.

The first part of the experiments was started immediately after the termination of irradiation period in order to have the greatest sensitivity for the relatively short-lived fission products and in order to simulate as closely as possible conditions following the hypothetical accident.

Each plate was tested in the following manner.

1. The plate was visually examined and photographed, using both Polaroid film and Kodak Tri-X professional film. The plate thickness was measured with a micrometer at 16 locations, the width at 3 locations, and the length at 2 locations.
2. The irradiated fuel plate was mounted on the quartz bed and then the bed was inserted into the quartz tube. And the tube was connected to the fission products collection system.
3. A stream of helium was passed through the tube and the collection system. Normally a flow rate of 100 ml/min was maintained.
4. The furnace was heated to the testing temperature. The temperature was maintained for 30 min.
5. The furnace and the fuel plate were allowed to cool.
6. After the fuel plate had cooled to approximately 100°C below the test temperature, the inlet petcock to the sample trap was closed, while the outlet one remained open. At this step, the flow of helium was diverted through the second fission product trap and backup trap. Then the liquid nitrogen bath was removed from the trap. As the sample trap was warmed up to the room temperature, the helium gas adsorbed on the charcoal passed through the down stream side of the system. Xenon losses were estimated to be negligible in this warm-up operation.
7. The sample trap was removed from the system and sealed. The used trap was moved from hot cell to the Ge(Li) detector and counted for 3 to 16 hours. Another new trap was installed. It was estimated that the counting error for Xe-133 or I-131 is less than about 10 % for the activity level of 10 pCi.
8. After the plate cooled to about 200°C, the fuel plate was removed, from the bed inspected, measured, and photographed as described in the Step 1.
9. The fuel plate was then placed back on the bed and the test was repeated at a higher temperature.

These tests were done at 400, 450, 500 and 550°C for the plate E-141, at 450, 500 and 550°C for the plate E-116, at 400, 450, 500 and 550°C for the plate E-101, and at 300, 400, 450, 500, 525 and 550°C for the plate E-114 respectively.

Subsequently, the minimum temperature at which rapid release occurred was determined for the plates E-141 and E-116. The plate was placed in the furnace and heated to 550°C. After that, the temperature was slowly increased at a rate of 1.1°C/min under the monitoring by the ionization chamber for fission product gas release.

For the plate E-141 the furnace was turned off when a rapid release of fission products was detected. The trapping of Xe-133 from the helium stream was continued for 1.2 hr, the furnace had been cooled to 222°C.
For the plate E-116, the plate was held for 30 minutes at the release-threshold temperature 561°C, and then the plate was allowed to cool.

Meltdown Experiment

In the second part of the experiment the plate E-114 was tested at higher temperatures than the temperature of the first rapid release of fission products. The equipment differed from that of the first part as follows: a caustic scrubber and a cold trap were placed in the line between the furnace tube and charcoal trap. The scrubber consisted of a 500 ml filter flask containing 300 ml of 50 % sodium hydroxide solution. The helium gas entered to the scrubber through a sintered glass and exited through the side arm of the flask, and passed through a cold trap before entering the charcoal trap.

The charcoal traps used in this part of the experiment were made of metal rather than glass and had valves in both inlet and outlet legs. This allowed safe handling of the traps with manipulator from the outside of the hot cell, because these traps were expected to hold appreciable quantities of radioactive gases.

The flow diagram of the equipment of part 2 is shown in Fig.1. and a photograph of the manifold section consisting of the sample traps and scrubber is shown in Photo. 2. Test was made at 565, 585, 600, 650 and 700°C. After holding at each temperature for 30 minutes the furnace was cooled to about 250°C and then the fuel plate was removed, inspected, and photographed.

The sample traps were transported in lead shielded cans to the Xe-133 Production Facility of the Isotope Department of ORNL. Here the xenon was transferred from the heated sample trap to a storage trap at liquid nitrogen temperature. During the process the gas was passed through ascarite to remove carbon dioxide, the principal impurity. The xenon was then condensed into glass ampules and taken to Analytical Division for analysis. Residual Xe-133 and other radioactive elements in the sample trap was measured by gamma-counting. Aliquots of the caustic solution were taken after each run and analyzed using a Ge(Li) counter.

After the 700°C test, the plate E-114 was taken to the Short-lived Fission Product Production Facility of the Isotope Department. Here the aluminum was dissolved in 6 M NaOH. This step was repeated. The residue was treated with 6 M HNO_3 three times to dissolve the residue completely. The principal gamma emitting isotopes in the solutions were determined by their gamma-ray measurements.[10] The uranium contents were determined by chemical analysis. The isotopic composition of the uranium in the first acid solution was determined by mass spectrometry.

Results and Discussion

The release of fission products from the fuel plate (UAl_x-Al) at temperatures below 550°C was found negligible except for the plate E-101 as

shown in Table 2. The observed values of activity of fission product release at temperature below 500°C were of the order of nCi per plate. All activity values are corrected to the value at the time of reactor shutdown. The amount of fission product activities per one gram of U-235 in the target after 260 day irradiation at 2 x 10^{14} n/cm^2sec were calculated as follows:[11] I-131: 13.9 Ci, Te-132: 18.8 Ci, I-132: 17.5 Ci, and Xe-133: 31.6 Ci. Some parts of the observed release of fission products may be caused by traces of uranium contamination on the surfaces of the fuel plate and/or natural uranium impurity existing in cladding material of 6061 aluminum alloy. The factor of ten higher release recorded for the plates E-141 and E-116 at the initial heatups may be caused by traces of uranium contamination on the surface of the plate by an initial desorption of gases.

Table 2. Fission Products Released from Samples

Run No.	Sample code	Furnace temp.(°C)	Amount released from specimen(nCi) Xe-133	I-131	Cs-137
A-1	E-141	400	89.2	1.5	0.03
A-2	E-141	450	2.2	3.1	0.03
A-3	E-141	500	7.9	12.1	0.04
A-4	E-141	550	5.8	8.2	0.01
B-1	E-101	400	8.3	0.4	0.05
B-2	E-101	450	3.2	15.6	0.03
B-3	E-101	500	9.4	7.9	0.03
B-4	E-101	550	$104x10^9$	$7x10^5$	$9x10^5$
C-1	E-116	450	83.6	4.9	0.03
C-2	E-116	500	6.9	1.1	0.02
C-3	E-116	550	2.3	38.8	0.02

Reynolds found that the release of fission product gases was negligible below the eutectic temperature of U-Al_4, 640°C, with the experiments using the lightly irradiated Al-20 wt% U alloys.[7] Graber et al.[8] reported that the fission gas release during the experiments on the dispersion fuels began at the solidus temperature (582°C) of the 6061 aluminum cladding by counting ^{85}Kr.

For the plate E-101 which has the thinnest cladding of all the plates (0.30 mm), a large amount of fission product release was observed at 550°C. The threshold temperature for rapid release of fission products was measured

to be 561±1°C for the plates E-141 and E-116 of which the cladding thickness were 0.39 mm. The plate E-114 was heated to 565°C in a single step from 550°C, and a large release was observed. This is consistent with a 561°C threshold temperature. In these experiments, it became clear that the fission products began to release at 561°C which is below the solidus temperature of cladding material.

No distortion of the plates was observed at temperatures up to 500°C. Warping was observed after the one test at 525°C and after all tests at 550°C. No blistering was observed at 550°C or lower temperature except for the plate E-101. Large blisters were observed every time after the thermal treatment in which a large fraction of fission product gases, such as xenon and iodine, were released. It can be said that the rapid release of fission products was caused by the occurrence of blistering. The blister occurred on the surface of the fuel plates was larger than that of the U-Al alloy fuels measured by Reinke.[4] The large blister observed on the surface of the cladding may be due to poor adhesion between the cladding material and the meat or to poor adhesion within the meat inself.[12]

The amounts of Xe-133 collected during the initial rapid release as the result of the occurrence of blistering of the cladding material are shown in Table 3 for each plate.

Table 3. Xenon-133 from the Initial Release

Sample code	Temperature (°C)	Xe-133 (Ci)	Other Radioactive Elements (mCi)
E-101	550	104	I-131: 0.7, Cs-134: 0.8, Cs-136: 0.5, Cs-137: 0.9,
E-116	561	19	I-131: 0.03
E-141	561	23	
E-114	565	47	

In order to investigate the mechanism for initial rapid release, ANL has resently carried out a metallographic examination at ORNL of one of the blistered plates. The surfaces of the blisters show numerous cracks. Preliminary results along metallography indicate[13] that incipient melting occurred at the grain boundaries along which the cracks formed. Based upon the present experiments alone, it is not possible to determine if incipient melting of the cladding is necessary for the release of fission products or if stresses produced during blistering are sufficient to cause cracks in the cladding. However, a recently-completed series of fission-product-release experiments on similar miniplates containing U_3O_8 and U_3Si dispersions showed that fission products were released when the plates blistered[14], even though

the blister-threshold temperature were as low as 500°C. In these cases the cracks which formed in the blistered cladding could not be attributed to melting. Of course, the limited data from these tests is not sufficient to show that formation of a blister will always be accompanied by a fission-product release. The temperature of the fuel plate and the size and shape of the blister could be important parameters related to the formation of cracks in the cladding. However, from the point of view of a safety analysis one must assume that a significant release of fission products may accompany blister formation. It is quite likely, considering the low uranium loading (1.3 - 1.4 gU/cm^3) and the relatively low burnup (0.5 - 0.7 x 10^{21} $fiss./cm^3$) of the plates tested by Graber, et al., that blistering did not occur below 582°C. If such were the case their data would not be inconsistent with the more resent data. Under this assumption, the following conclusion can be reached on the basis of all fission-product-release threshold-temperature data: The threshold temperature for the first significant release of fission products from dispersion fuels, clad in 6061-aluminum alloy, is the lower of the blister-threshold temperature or the cladding solidus temperature.

Table 4 shows the amounts of I-131 and Cs-137 collected in the caustic scrubber solution for the plate E-114 at temperatures above that for initial rapid release. The amount of iodine released increased with the temperature. Even at 700°C only a small fraction (5.5 x 10^{-3} %) of the total I-131 was released as shown in Fig. 2. Since cesium has a low vapor pressure at these temperature range, the rate of release of cesium was slower than that of the inert gases. The amount of released Cs-137 at 700°C was of the order of 10^{-3} % to the overall Cs-137 activity in the plate. Gamma scans of the charcoal traps showed that a small fraction of the iodine passed through the caustic scrubber. Some iodine might also be deposited on the surfaces of the furnace tube or the glass tube connecting the furnace tube to the scrubber.

The amounts and the relative values of Xe-133 released during the high temperature runs for the plate E-114 are also given in Table 4.

Table 4. Fission Products Released from Meltdown Experiment on Plate E-114

Temp. (°C)	Xe-133 Trapped in Charcoal (Ci)	(%)	I-131 Trapped in Scrubber (dps)	Cs-137 Trapped in Scrubber (dps)
565	47	20.4	$8.5x10^4$ (2.3 μCi)	$5.2x10^3$
585	116	50.2	$4.1x10^6$ (0.11 mCi)	$3.4x10^4$
600	7	3.0	$2.0x10^7$ (0.54 mCi)	$6.2x10^3$
650	57	24.7	$2.0x10^7$ (0.54 mCi)	$8.2x10^5$
700	4	1.7	$3.0x10^7$ (0.81 mCi)	$1.3x10^5$

As Discussed earlier the first rapid release of fission product gases occurred at or below 565°C and was accompanied by blistering of the plate, as shown in Photo. 3. Another large release occurred at or below 585°C, probably upon reaching the solidus temperature of the cladding (Photo. 4). The much larger release at 585°C than at 565°C indicates that the voids containing fission gases were not totally interconnected and that only gases from the immediate vicinity of the blisters participated in the initial release. Another large release of fission gas occurred at or below 650°C (Photo. 5) and very probably happened at the U-AL alloy eutectic temperature, 640°C. At the 700°C temperature most of the ^{133}Xe had probably been released but most of the ^{131}I was still in the plate. From these results it can be concluded that for highly-loaded UAl_x-Al fuel up to 70% of the fission gas resides in the aluminum matrix region and is available for release upon cladding melting.

A cracking of the cladding was observed after the 650 °C experiment as shown in Photo. 5. At the 700 °C experiment, crumples were observed on the surface of the plate E-114 which indicated the melt of the plate as shown in Photo. 6.

Table 5 shows the total amounts of uranium and principal fission products remaining in the plate E-114 as determined by chemical analysis. The table shows both the measured values at the time of analysis and the corrected values corresponding to the reactor shutdown time (07/10/81).

The isotopic composition of the uranium in the plate E-114 after irradiation is given in Table 6. The 0.980 g of U-235 remaining in the plate indicates a burnup of 63% and a fission density of $1.06 \times 10^{21}/cm^3$.

Table 5. Fission Products in Plate E-114

Sample	Zr-95 (mCi)	Nb-95 (mCi)	Ru-103 (mCi)	Ce-141 (mCi)	Ce-144 (mCi)	Cs-134 (mCi)	Cs-137 (mCi)	Ba-140 (mCi)	La-140 (mCi)	U (g)
Acid 1										
09/03/81	1.17×10^4	7840	877	873	9810			2907	3047	0.387
07/10/81	2.12×10^4		2308	2.82×10^4	1.22×10^4			5.73×10^4		
Acid 2										
09/18/81	3.78×10^4	4334	517	4770	7065			522	562	0.154
07/10/81	8.08×10^4		1773	2.12×10^4	8379			2.32×10^4		
Acid 3										
09/18/81	8.09×10^4	4.74×10^4	6250	3.66×10^4	5355			3961	4488	4.22
07/10/81	1.73×10^5		2.14×10^4	1.63×10^5	6351			1.76×10^5		
Base 1										
09/03/81	880	589	294	722	836	722	2980	974	984	
07/10/81	1792		775	2333	956	760	2888	1.70×10^4		
Base 2										
09/18/81	115	160	12	19.6	29	27	9.4	11	12	
07/10/81	245		41	87	34	29	9.4	491		
Total										
07/10/81	2.77×10^5		2.63×10^4	2.15×10^5	$2.79\ 10^4$	789	2897	2.73×10^5		4.76

Table 6. Isotopic Composition of Uranium in Plate E-114

Isotope Mass	Percent of Isotope
234	0.32 ± 0.01
235	20.59 ± 0.03
236	5.41 ± 0.01
238	74.0 ± 0.03

Conclusions

These experiments using the fuel plates of burnup of about 60 % established that the threshold temperature for fission product release from U-Al_x-Al dispersion plate-type fuel, clad in 6061-Aluminum alloy, is 561±1°C, below the solidus temperature of the cladding.

The first significant release of fission product gas apparently occurs by blister formation. The second release causes by melting of cladding at the solidus temperature of 582°C. The last massive release occurs at the eutectic temperature (640°C) of U-Al_x.

The amounts of released iodine-131 and cesium-137 are of the order of 10^{-3} % of the total iodine-131 and cesium-137 in the plate respectively.

The release threshold temperature appears to be dependent on the burnup , fuel material and the cladding thickness.

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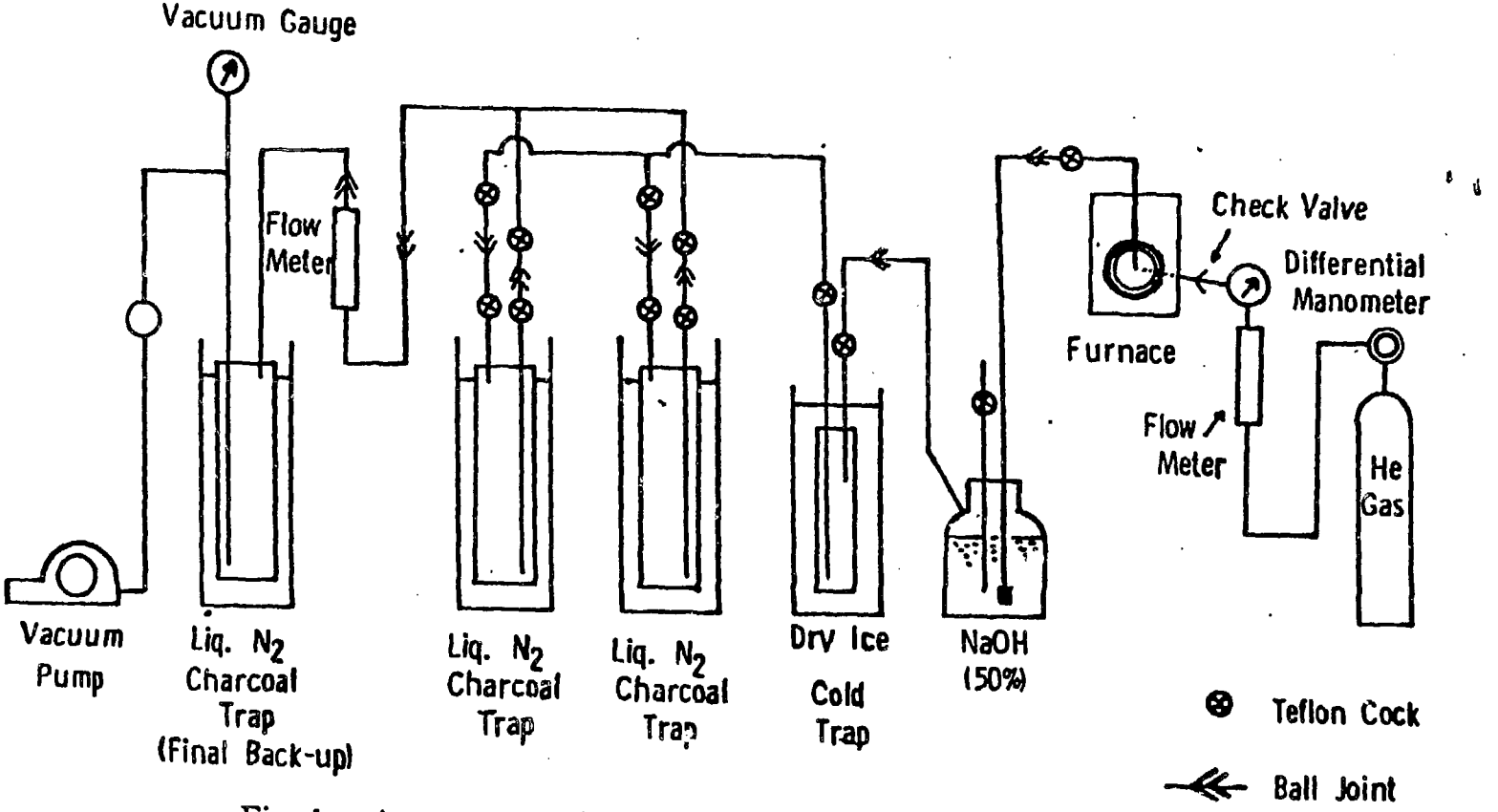

Fig. 1. Apparatus with metal traps for meltdown experiment.

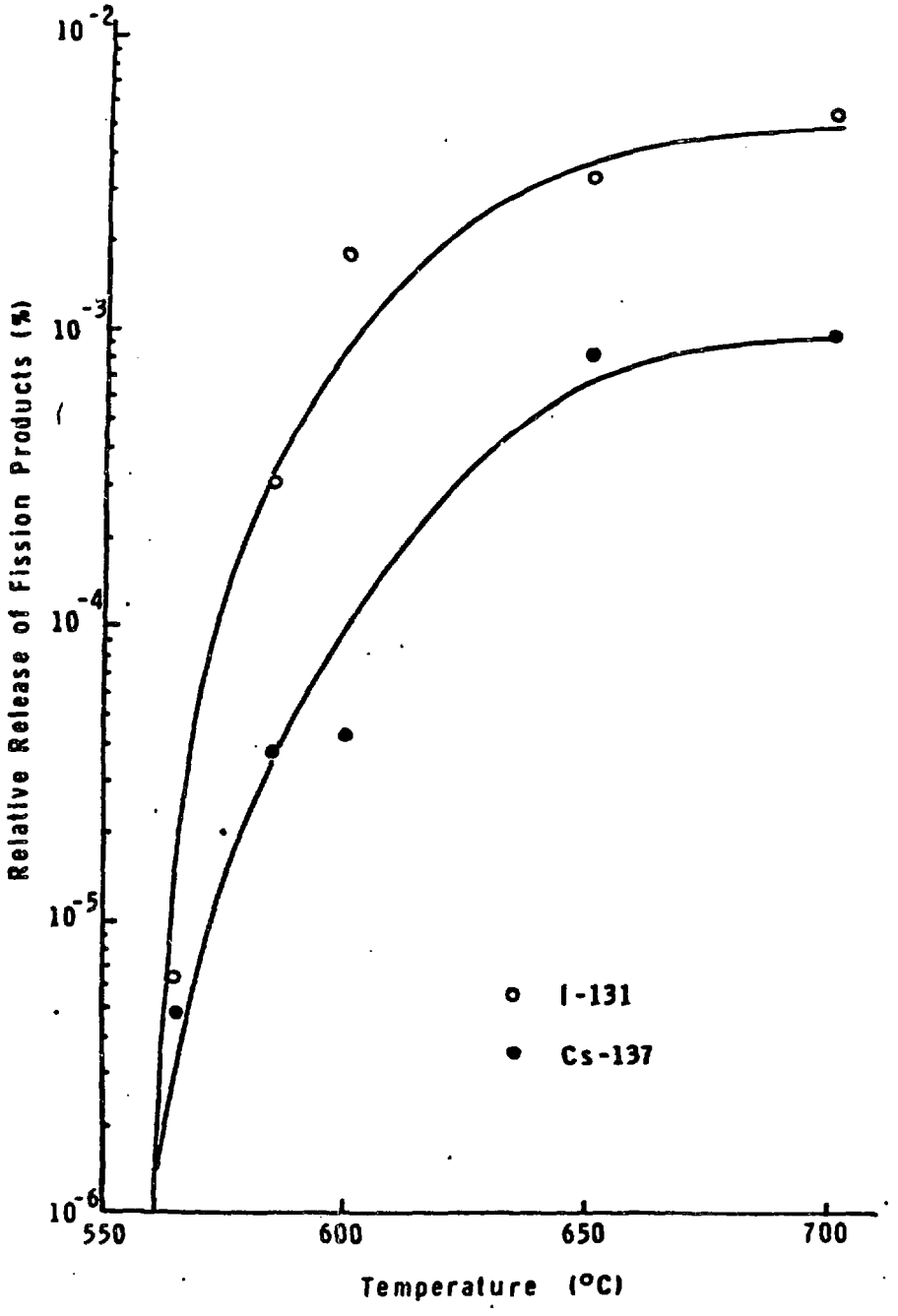

Fig. 2. Cumulative amounts of I-131 and Cs-137 released from the plate E-114 after keeping for 30 min. at various temperatures.

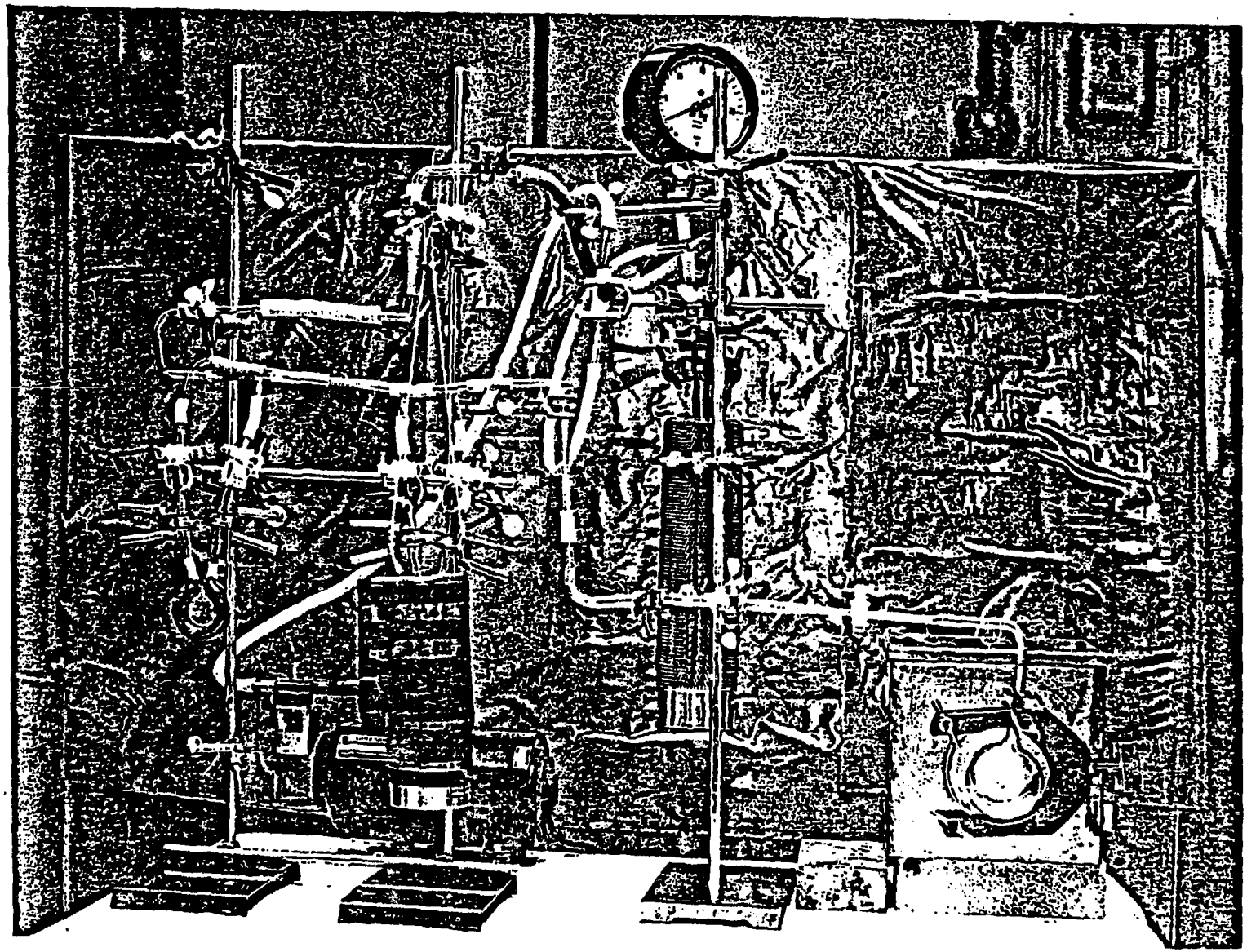

Photo. 1 Equipment used for trapping of fission product gases.

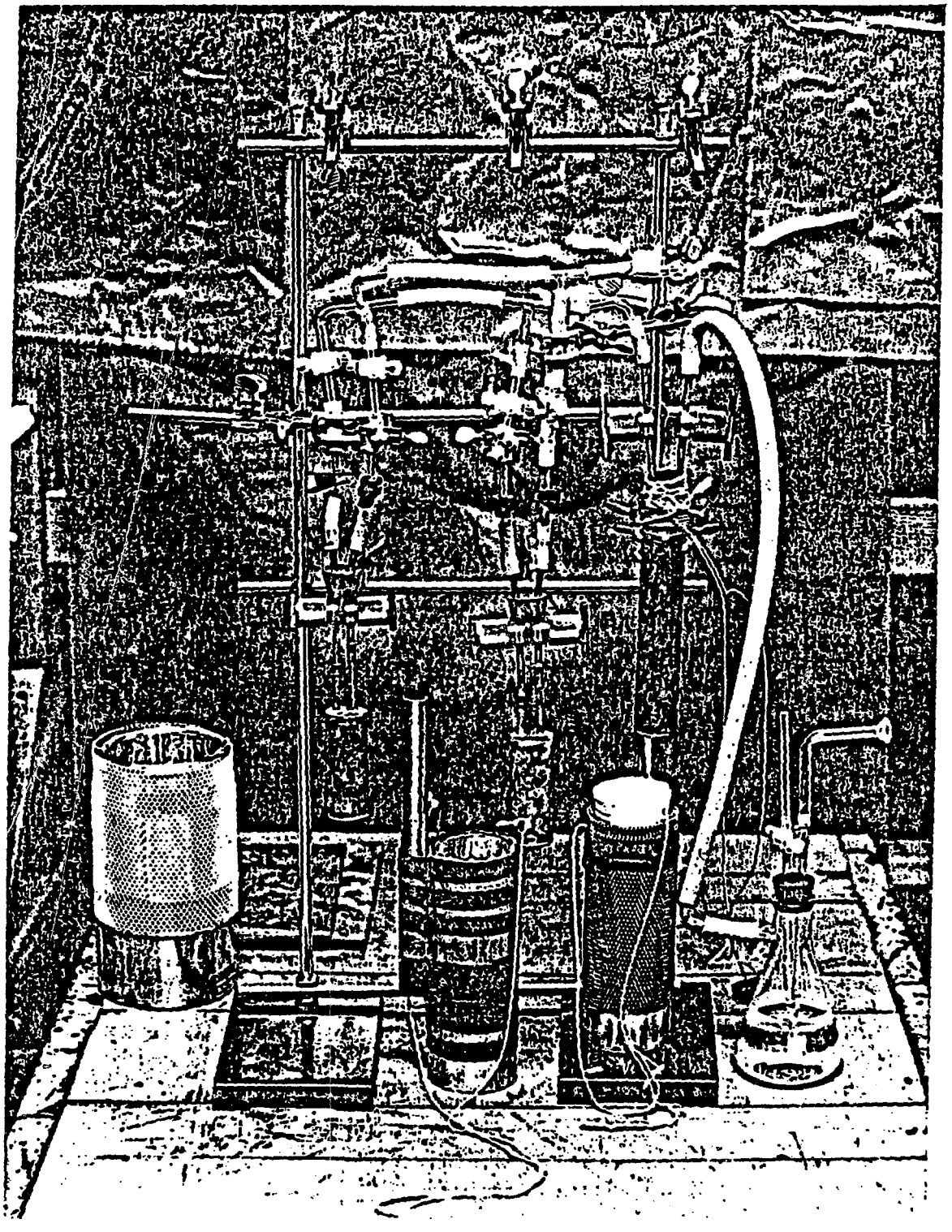

Photo. 2 Equipment used for meltdown experiment consisting of the metal traps and scrubber solution.

Photo. 3. Plate E-114 after heating up to 565°C.

Photo. 4. Plate E-114 after heating up to 585 °C.

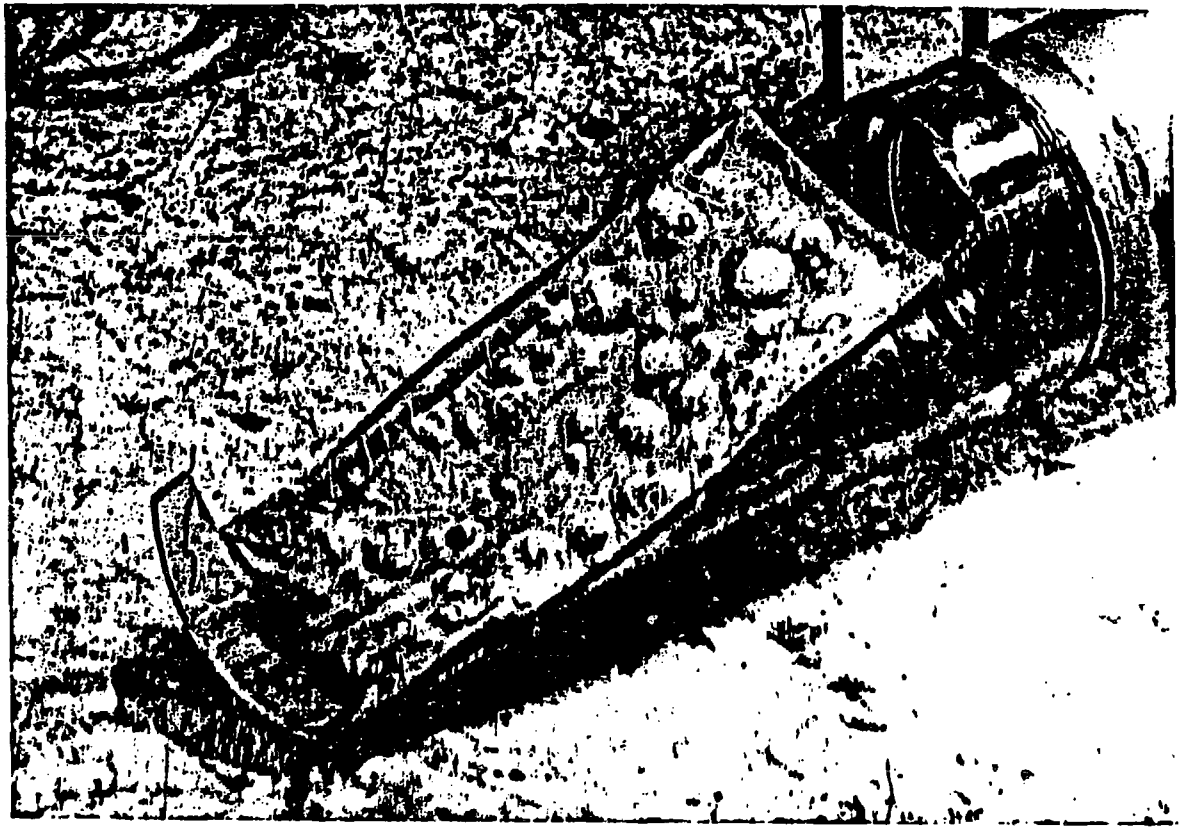

Photo. 5. Plate E-114 after heating up to 650 °C.

Photo. 6. Plate E-114 after heating up to 700 °C.

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