HW-72321

UC-41, Health and Safety
(TID-4500, 17th Ed.)

FISSION PRODUCT RELEASE FROM URANIUM--EFFECT OF IRRADIATION LEVEL

By

R. K. Hilliard
D. L. Reid
Chemical Effluents Technology
Chemical Research and Development Operation
Hanford Laboratories

June 20, 1962

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1)-1350 between the
Atomic Energy Commission and General Electric Company

Printed by/for the U. S. Atomic Energy Commission

Printed in USA. Price $1.25. Available from the
Office of Technical Services
Department of Commerce
Washington 25, D.C.

ABSTRACT

An experimental study was made to determine the influence of burnup on the fractional release of fission products from uranium metal at elevated temperatures. Neutron irradiation levels were varied from 2×10^{14} nvt (6.7×10^{-4} MWD/T) to 4×10^{20} nvt (1340 MWD/T) while all other conditions remained constant. Metal temperatures explored were 1000, 1200, and 1440 C. Bare uranium cylinders weighing approximately 12 grams were heated out-of-reactor and the liberated fractions of ten fission products plus uranium, plutonium and neptunium were measured. The variation of the fractional releases of iodine, xenon and cesium with irradiation level was shown, as was the invariance of the release of certain other elements with burnup. The uranium oxidation rate was found to increase with irradiation levels above 10^{18} nvt.

TABLE OF CONTENTS

Page

FISSION PRODUCT RELEASE FROM URANIUM -- THE EFFECT OF IRRADIATION LEVEL

INTRODUCTION

An important consideration in appraising the consequences from hypothesized accidents with nuclear fuels is the fractional release of the radionuclide inventory of the fuel under prevailing conditions.

The earliest estimators(1, 2) assumed that 100 percent of all the fission products would be released. Although this was believed to be ultra conservative, no experimental evidence was available to justify the use of lower values. In the past few years much work was done on the problem at several installations, and the resultant information showed that the bulk of the total fission products would remain in the fuel.(3-13) Most of this experimental work was performed under conditions which would impart a degree of conservatism when the data were applied to reactor conditions. Thus, extreme temperatures, the most destructive type of atmosphere, prolonged heating periods, specimens with high surface area to weight ratios and unclad fuels were used to determine the maximum fission product release.

One condition was generally employed in the experimental work, however, which might have given low release values. This was the irradiation level of the test specimens. For hazard evaluations, fuel involved in a reactor or fuels reprocessing plant accident must be defined as being highly irradiated, whereas most of the experimenters used specimens irradiated to very low levels in order to avoid the difficulties of handling highly radioactive materials. Results of a few tests at Oak Ridge National Laboratory indicated that more highly irradiated fuels released greater proportions of some of the fission product elements.(3, 4) If the magnitude of this effect is sufficiently large, presently applied fractional-release estimates might require adjustment upward.

An investigation was made at Hanford Atomic Products Operation which had as its primary objective the determination of the extent to which burnup influences the release of fission products from metallic uranium. The results of this study constitute the basis of this report.

SUMMARY

The fraction of ten key fission product elements plus plutonium, uranium and neptunium released from metallic uranium during out-of-reactor heating in air was measured at various specimen irradiation levels. Tests were conducted at temperatures above and below the uranium melting point of 1132 C.

For specimens above the uranium melting point, an increase in the irradiation level from 2×10^{14} nvt (6.7×10^{-4} MWD/T uranium) to 4×10^{20} nvt (1340 MWD/T) resulted in:

(1) an increase in rare gas release from about 70 to 99.8 percent,
(2) an increase in iodine release from about 60 to 95 percent,
(3) an increase in cesium release from about 30 to 80 percent,
(4) an increase in the release of ruthenium and molybdenum from <0.1 to about 5 percent, and
(5) no discernable effect on the release of tellurium (65 percent), strontium and barium (0.5 percent), or the nonvolatile elements, zirconium, cerium, plutonium, neptunium and uranium (<0.1 percent).

Tests with specimens below the melting point generally gave similar trends for the influence of irradiation level.

Higher irradiation levels caused a more prompt release of the gaseous fission products. Xenon was released almost quantitatively when the highly irradiated metal melted, while trace-level specimens continued to release xenon throughout the heating period.

The average uranium oxidation rate increased with irradiation levels above 10^{18} nvt (about 3 MWD/T); the rate was 30 percent greater at 4×10^{20} nvt than at 10^{18} nvt.

The formation of gas bubbles during irradiation is proposed as an explanation of the increased fractional release of some fission products, the prompt release of volatiles, and the higher uranium oxidation with increasing irradiation level.

EXPERIMENTAL CONDITIONS

General Approach

Since the specific objective of the investigation was to determine the effect of fuel burnup on the release of key fission products from uranium, an effort was made to fix all variables except irradiation level. Temperature was shown by earlier tests to be an important factor for the release of most fission products from uranium irradiated to trace levels.(11, 12) The study was divided into three series conducted at temperatures of 1000, 1200 and 1440 C. The majority of the tests was arbitrarily made at 1200 C, 68 C above the uranium melting point.

The general approach was to heat the test specimen rapidly to the desired temperature in flowing helium, then to keep an isothermal condition while air flowed over the specimen for a specific period period of time, after which helium atmosphere was restored and the specimen was allowed to cool rapidly. The furnace effluent was scrubbed and filtered during the test to remove all radioactive material. These traps and filters, with their interconnecting lines, were decontaminated later to recover the released material.

After cooling, the residue was transferred to a dissolver where preferential dissolving allowed determination of uranium oxidation and residual fission products. Initially, duplicate uranium specimens were dissolved to provide material balances. Thus, three solutions were analyzed radiochemically: V, the volatile fraction which consisted of all solutions from off-gas traps and lines; R, the residual fraction; and D, the duplicate solution which represented the total original fission product inventory. After determining that the sum of the residual and volatile fractions approximated the original fission product inventories, the use of duplicate samples was discontinued and the following equation was used for the calculation of all release fractions for all tests:

$$\text{Fractional Release} = \frac{V}{R + V}$$

An air atmosphere was selected because previous tests resulted in much greater releases of most fission products in air than in steam or helium;(13) a burnup effect should not depend on the type of atmosphere used.

Test Specimens

The reactor grade uranium specimens were 1/4-inch diameter by 3/4-inch long cylinders weighing 11.7 ± 0.3 g. The cylinders were etched with concentrated nitric acid, hand polished with 4/0 polishing paper, weighed and sealed under a helium atmosphere into aluminum holders. Specimens which were not beta heat-treated after swaging exhibited severe growth at irradiation levels above 10^{19} nvt. An extra heat treatment eliminated the growth problem, but surface irregularities persisted at levels above 10^{20} nvt.

Usually at least two specimens were irradiated in the same holder so that a duplicate could be dissolved and analyzed for fission product content. In some of the tests 1/4-inch diameter by 1/8-inch thick uranium disks were used for the duplicate.

The specimens were irradiated in a reactor to predetermined levels and then were allowed to decay for a suitable period before testing. The specimens exposed to the lowest levels were decayed 5 days; the decay period for the longer exposed specimens was about 30 days. A maximum exposure of 4×10^{20} nvt was attained.

For the purpose of comparing the irradiation level expressed in other ways, the following relationships can be used:

$$\text{MWD/T} = (3.4 \times 10^{-18})(\text{nvt})$$

$$\text{Atom Percent Burnup} = (4 \times 10^{-22})(\text{nvt})$$

where MWD/T is megawatt days per ton of natural uranium, atom percent burnup is the burnup of all fertile plus fissionable atoms, and nvt is the time-integrated thermal neutron flux. These conversion expressions are essentially correct for the conditions used in these tests even though corrections for fast flux fission and flux depression are not included. Estimated exposures by both radiochemical measurements and by calculations based on reactor parameters agreed to within 20 percent.

Experimental Equipment

A small shielded cell was constructed for use in these studies. Its interior stainless steel lining measured 24 x 24 x 28 inches high. Stacked lead bricks provided eight inches of shielding. Two ball-type manipulators were located on the front face. A removable top section contained two holes for portable, vertical finger-grip manipulators used for loading the furnace and remote sampling.

The furnace, two caustic scrubbers and a dissolver were located inside the cell. The off-gas line from the scrubbers led into a "Junior Cave" where cold traps, an absolute filter and a charcoal bed were located. A photograph of the cell and "Junior Cave" is shown in Figure 1. The cold traps were glass columns filled with glass beads kept at -70 C by a dry ice-isopropanol slurry. The Millipore filter, type AA, 0.8 μ pore size, was supported by a Pyrex holder. The charcoal bed, which contained 75 grams of activated coconut charcoal of 12-14 mesh, was also cooled to -70 C by dry ice-isopropanol. A steam jet provided a vacuum source for drawing dried room air through the system. A complete flow diagram is shown in Figure 2.

The furnace was heated inductively by a 15-kw motor-generator of 10,000 cps frequency. The water-cooled copper work coil entered the cell through a hole in the lead shielding. The furnace proper, made of 2-inch ID fused quartz, was oriented vertically within the work coil. Figure 3 shows a sketch of the furnace arrangement. The alumina crucible rested on the internal quartz thermowell, thus giving close proximity of the uranium specimen to the thermocouple junction.

Air or helium entered the furnace through the top and exhausted through the internal dip leg which extended into the crucible to within 1/2-inch

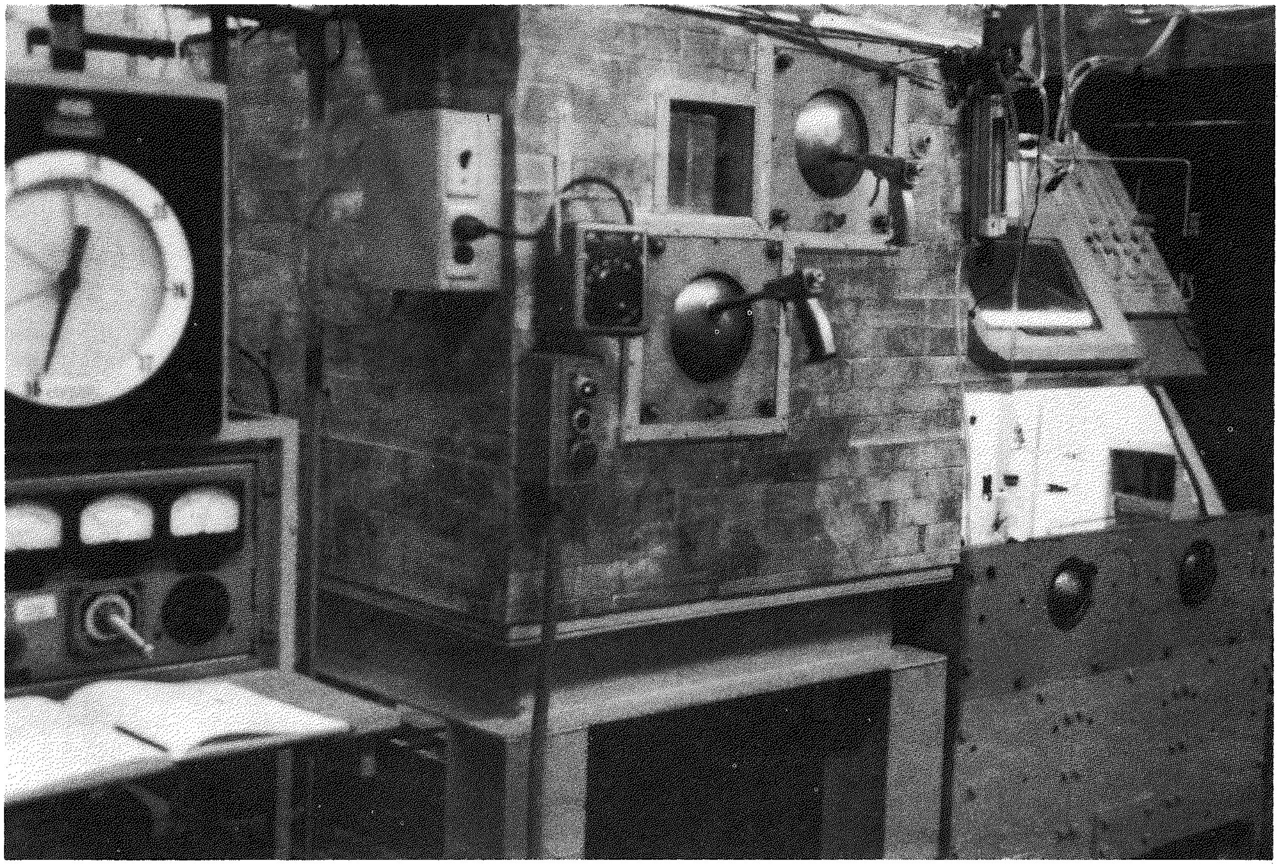

FIGURE 1

Cell and Junior Cave

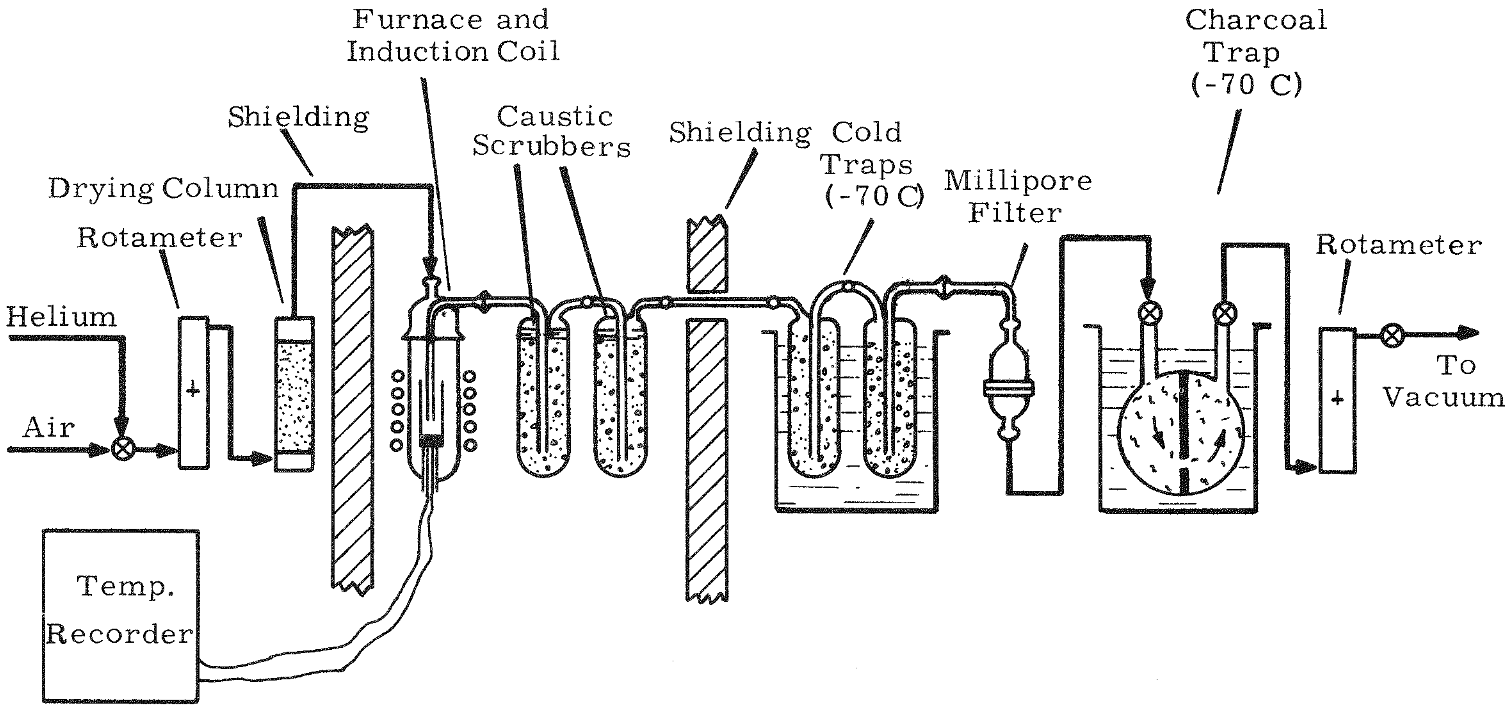

FIGURE 2

Flow Diagram of Fission Product Release Test Apparatus

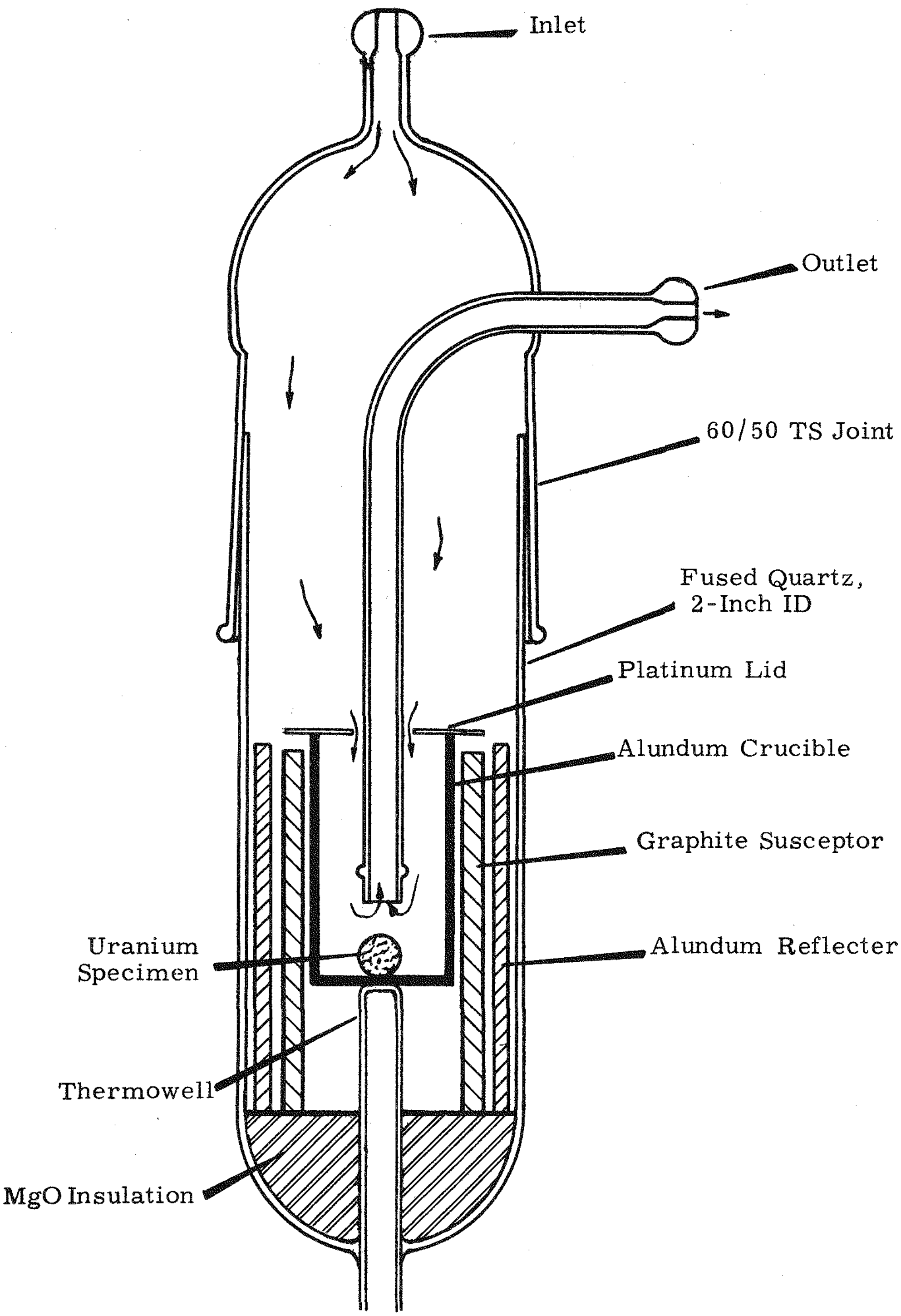

FIGURE 3
Furnace Arrangement

of the bottom. A loose fitting platinum lid semi-sealed the crucible and minimized counter-flow of air and evolved fission products from the crucible. In practice this scheme was only partially successful and the quartz furnace walls, graphite susceptor and alumina reflecter were invariably contaminated to a relatively low level.

The caustic scrubbers were Pyrex cylinders containing glass beads to aid in breaking up the gas bubbles. The first caustic scrubber contained 10 percent sodium hydroxide solution as the absorber; the second contained 30 percent sodium hydroxide.

Air flow was measured by rotameters at the furnace inlet and charcoal trap outlet. A three-way solenoid valve was used to switch from helium to air atmospheres. Both gases were dried by a desiccant. Pressure within the furnace was essentially atmospheric. Air and helium gas flow rates were 2300 cc/min (STP) in all tests.

Two dissolver models were used. Duplicate (unheated) specimens were dissolved in a stainless steel dissolver. The oxidized residues of the test specimens were dissolved in a heavy-walled Pyrex vessel suitable for remote operation. Electric mantles provided the heating for both dissolvers. In both systems the radioactive content of the off-gas from the dissolver was determined, but only the rare gases were released from the nitric acid solution to any significant extent. Figures 4 and 5 are photographs of the dissolvers.

The temperature was measured by a platinum-sheathed thermocouple composed of Pt to Pt-13 percent Rh. The thermocouple was spring loaded to assure contact with the end of the quartz thermowell on which the crucible rested. The temperature indicated by a thermocouple at the position of the test specimens was 5 C higher than that recorded at the thermowell position and appropriate allowances were made during tests. A potentiometer was used during each test to confirm the recorder reading.

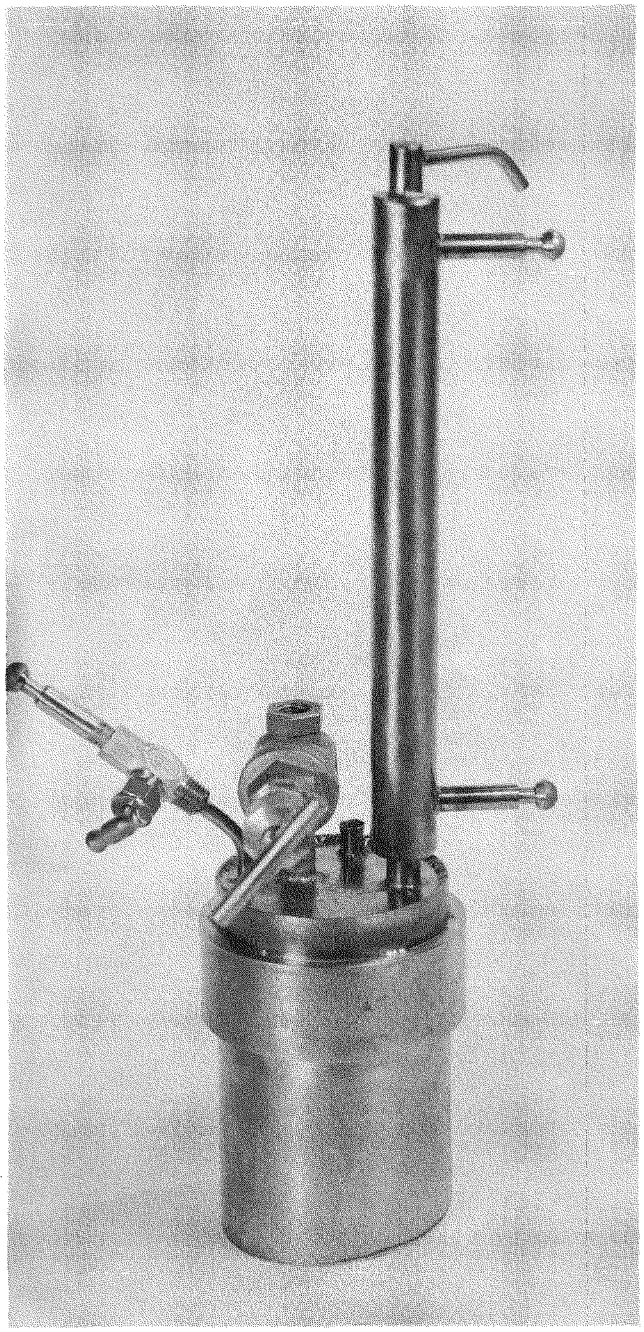

FIGURE 4

Stainless Steel Dissolver

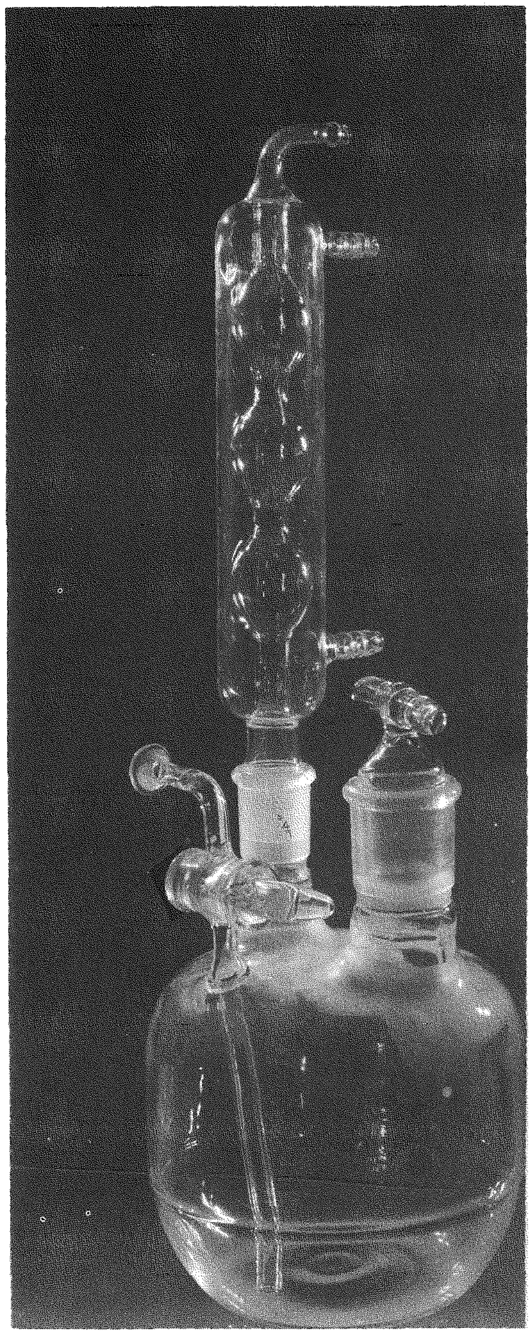

FIGURE 5

Residue Dissolver

Sample Preparation and Analysis

Solutions of the volatile, residual and duplicate samples were prepared in a manner suitable for radiochemical analysis. The nitric acid dissolver solutions were merely stirred, purged of rare gases, sampled and an aliquot submitted for analysis.* Preparation of the "volatile" samples required considerably more effort and care. Release was defined as the radioactive material found downstream of the crucible. The Millipore filter membranes were dissolved in hot 5 percent sodium hydroxide. In most of the tests the furnace lid and dip-leg washings were combined with those from the cold traps. The acidified filter solution was usually analyzed separately, as were the solutions from the caustic scrubbers. Hydrofluoric acid backwashes were found to decontaminate the glassware effectively. Wash solutions of the connecting lines were added to the adjacent trap solutions. In many cases, small aliquots were taken to reduce the dose rate to analytical personnel. Polyethylene bottles and stoppers were used for sample containers.

Uranium Oxidation Measurements

Experiments at Oak Ridge, Harwell and Hanford established that the release of volatile fission products from trace-irradiated uranium is proportional to the percent of the uranium oxidized.(4, 8, 13) Therefore, the experimental work was planned to duplicate all conditions which might affect the oxidation rate of uranium. Thus, the isothermal heating period, furnace configuration, specimen size, air flow rate, and manner of heating and cooling were the same in all tests for any particular temperature. Also, in order to give measureable quantities of all fission products in the off-gas system, rather severe conditions were set. Seventy percent oxidation of the uranium specimen was selected on the basis of trace-level studies. Test series at both lower and higher oxidations would have been advantageous, but would have increased greatly the number of tests. Therefore, preliminary tests using unirradiated uranium specimens were made in the vertical furnace (Figure 3) to establish oxidation behavior. It was found that 45

---

*Radiochemical analyses were performed by the Chemical Analysis Operation, Analytical Laboratories, Hanford Laboratories Operation.

minutes at 1000 C, 24 minutes at 1200 C, and 10 minutes at 1440 C gave 68 ± 4 percent oxidation as measured by metal weight difference. These results compared closely with those obtained in the horizontal resistance furnace apparatus used in previous trace-level tests.(13)

A chemical method for determining the percent of uranium oxidized was devised for use with the more highly irradiated specimens. The method was simply a two-step dissolution in concentrated nitric acid. Room temperature acid dissolved the oxide rapidly but attacked the remaining metal only slightly. After fifteen minutes at ambient temperature, the solution was stirred, purged of rare gases and an aliquot was taken. Then the acid was heated to dissolve the metal. The uranium content of each solution was determined by the controlled potential coulometric method to within 2 percent. Results using this method averaged 3.5 percent higher than by the weight-difference method for unirradiated specimens. Therefore, a correction was applied to the chemically determined values for the irradiated specimens.

Rare Gas Measurements

The fractional release of fission product rare gases was determined by comparing the counting rates of the charcoal traps. No attempt was made for absolute measurements, but the relative radioactive contents were determined by counting the gamma rays emitted through the stainless steel walls of the traps. Figure 6 shows the apparatus used. The traps were held in a jig at reproducible distances from the NaI (thallium activated) crystal. The crystal was 1-mm thick by 1-1/2 inches diameter and was mounted on a standard photomultiplier tube which fed through a single-channel analyzer to a decade scaler. Integral counts were taken with discrimination against energies <40 kev.

Use of the thin crystal and discrimination at 40 kev limited the detection range to 40-110 kev. Thus, the fission gas xenon-133, with its 81 kev gamma energy, was detected with reasonable efficiency, while interference from contaminants was greatly reduced. The only contaminant that could significantly affect the measurements was iodine-131. This isotope

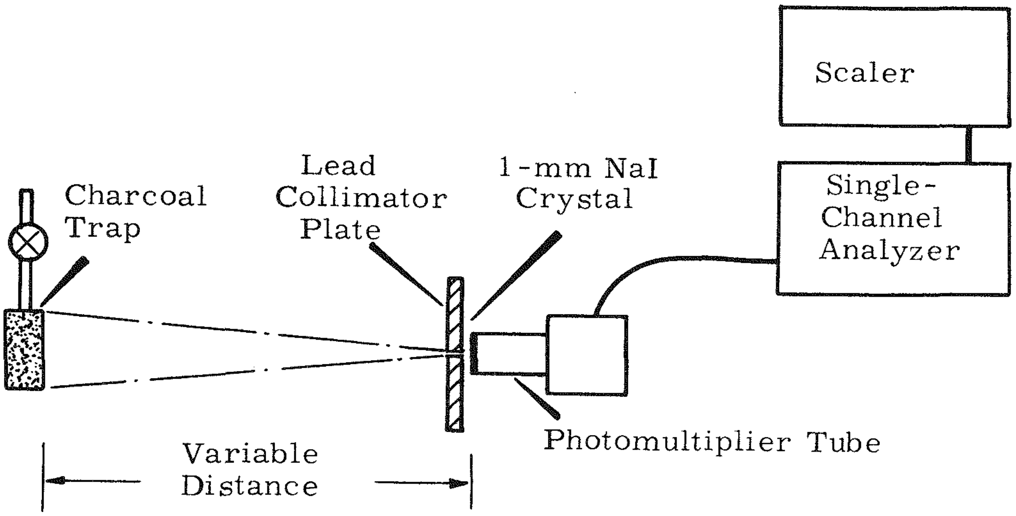

FIGURE 6

Xenon-133 Counting System

was found to be present in some of the charcoal traps, usually contributing <2 percent to the counting rate of the off-gas traps. Suitable corrections for this impurity were determined after driving off the xenon by heating the traps to 100 C and recounting.

The xenon-133 content of the uranium specimens ranged from a theoretical value of 10^7 d/m to 10^{11} d/m. Also, the off-gas trap for the higher irradiated specimens collected up to 99.9 percent of the total, giving it a counting rate 1000 times greater than the trap which collected the residual xenon during acid dissolution.

With trap activities varying by factors up to 10^6, a flexible counting system was required to provide reliable counting rates at all levels. This was accomplished by simultaneously increasing the distance between source and scintillation crystal and reducing the hole diameter in the collimator plate or vice versa as the irradiation levels of the specimens increased or decreased, respectively. All traps of a single test were counted under identical conditions.

RESULTS

The results of the experimental work are presented in tabular form in Appendix A. The tests are not listed in numerical order, but rather by increasing irradiation levels within each temperature group.

The wide range of radioisotope concentrations in the sample spectrum and the related decontamination factors required to produce reliable data caused many analytical problems. Methods used to analyze samples from slightly irradiated specimens were not suitable for samples from highly irradiated uranium. Analytical methods were improved as the work progressed; however, some data were lost and some of known low confidence value were produced. The relative method of calculating release fractions tended to normalize the data.

Effect of Irradiation Level on the Fractional Release of Fission Products

To simplify the presentation and discussion of results, the thirteen elements investigated are grouped into four classes (Table I) by relative volatilities. The "volatile oxide" group consists of those elements which are more volatile as the oxide than in elemental form. Tellurium, although meeting this description, is classed as "volatile" because of its high release rate. It should be emphasized again that the magnitude of the percentage release is a function of the experimental conditions used in these particular tests. A different choice of temperature, time or atmosphere would have resulted in considerably different releases. The major interest in this study was in the trends caused by variations of burnup.

TABLE I

CLASSIFICATION OF FISSION PRODUCT ELEMENTS ACCORDING TO VOLATILITIES

Class	Elements	Range of Release
Volatile	Xe, I, Cs, Te	20-100 %
Slightly Volatile	Sr, Ba	<1 %
Nonvolatile	Zr, Ce, Np, Pu, U	<0.1 %
Volatile Oxide	Ru, Mo	0.01-5 %

Volatile Elements

The effect of irradiation level on the release of elements in the "volatile" group is illustrated by Figures 7, 8, 9 and 10 for xenon, iodine, cesium and tellurium, respectively. The percent release is plotted as a function of the logarithm of the integrated thermal neutron flux (nvt). The fitted curves show an increasing release with increasing irradiation level for xenon, iodine and cesium. For the 70 percent uranium oxidation conditions of these tests, the effect of increasing the (nvt) by 10^6 was to increase the percent release of xenon by a factor of 1.4, iodine by 1.6 and cesium by 2.7. The scatter pattern of the tellurium data prevented the determination of any burnup effect (Figure 10). The reversion of tellurium release with increasing temperature is a function of the instability of the volatile tellurium oxide at high temperatures; the free energy of TeO_2 is zero at 1430 C.(14)

Slightly Volatile Elements

Strontium and barium were released to a much lesser extent than the elements of the volatile group but to a significantly greater extent than those of the nonvolatile group. The releases are plotted as functions of the irradiation levels in Figures 11 and 12 for strontium and barium, respectively. No trend due to irradiation level is apparent.

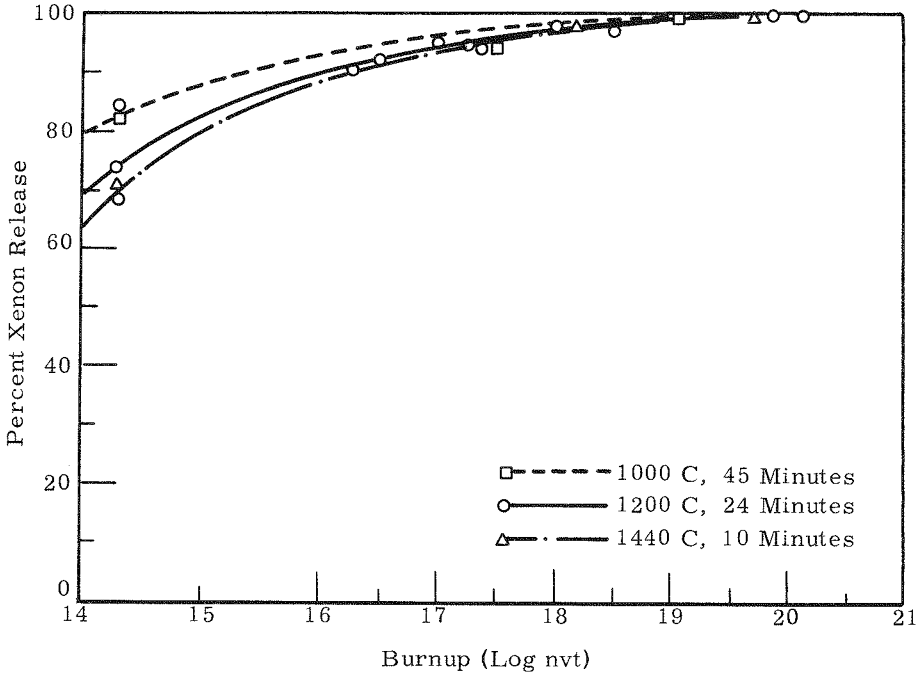

FIGURE 7

Release of Xenon as a Function of Burnup

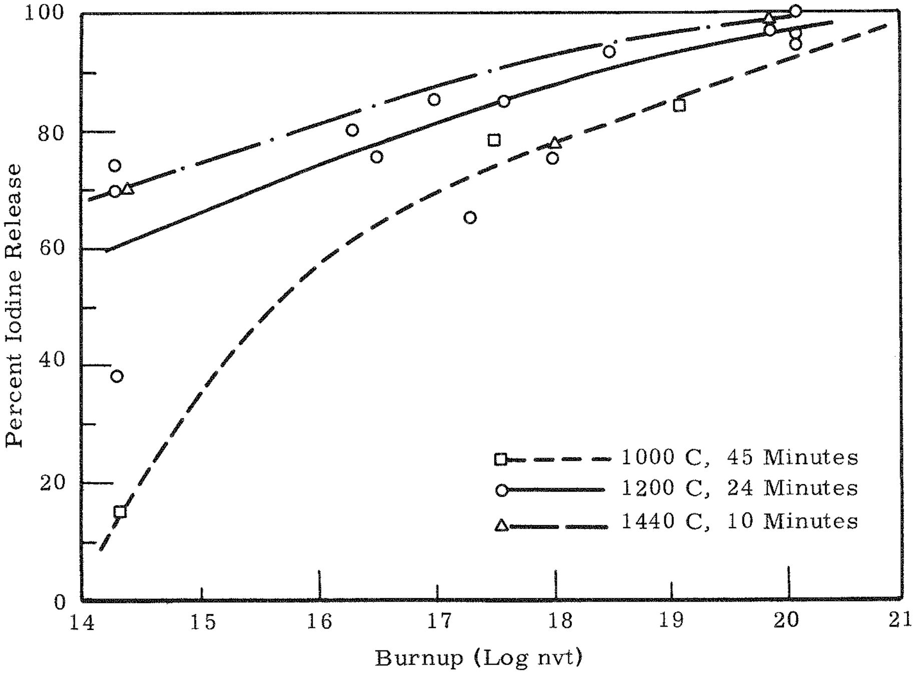

FIGURE 8

Release of Iodine as a Function of Burnup

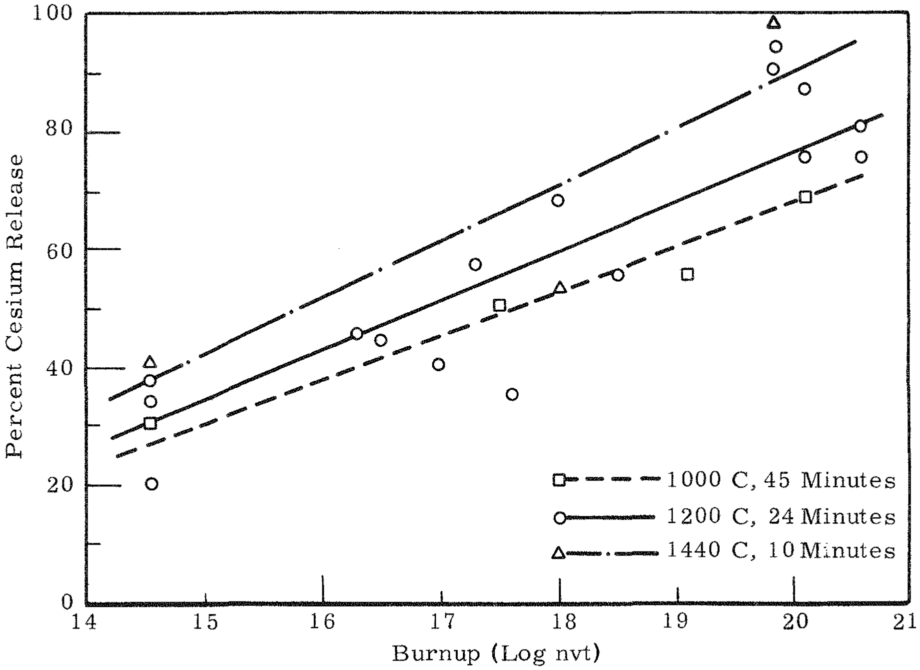

FIGURE 9

Release of Cesium as a Function of Burnup

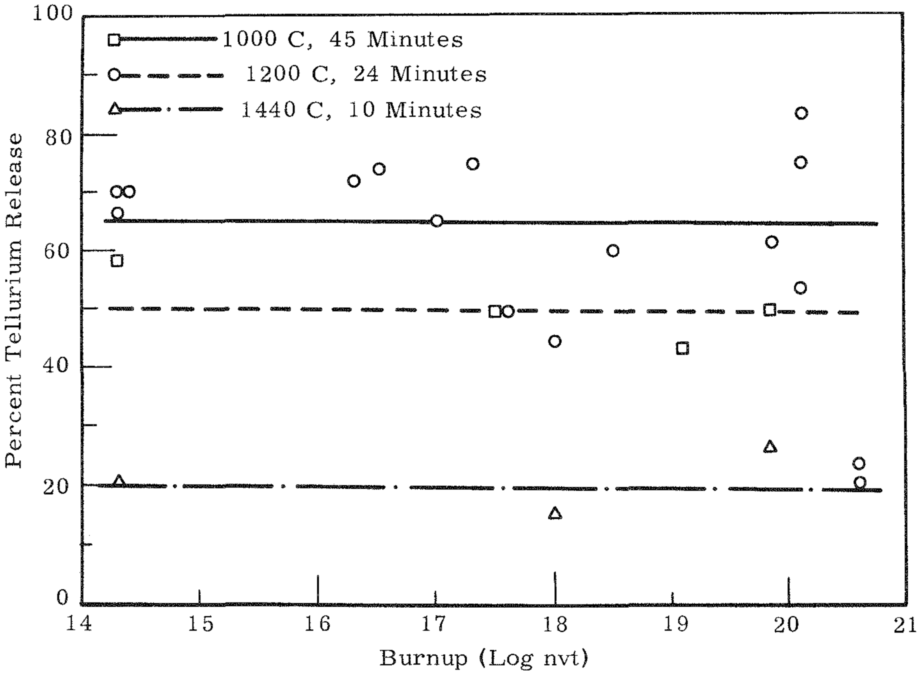

FIGURE 10

Release of Tellurium as a Function of Burnup

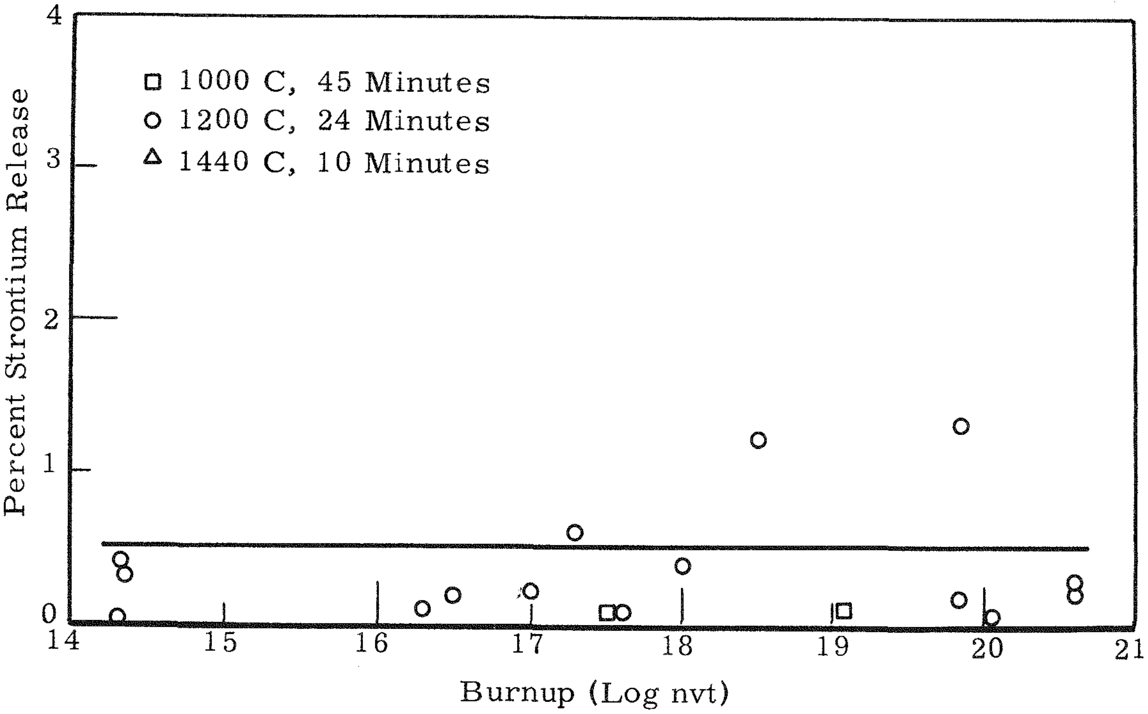

FIGURE 11

Release of Strontium as a Function of Burnup

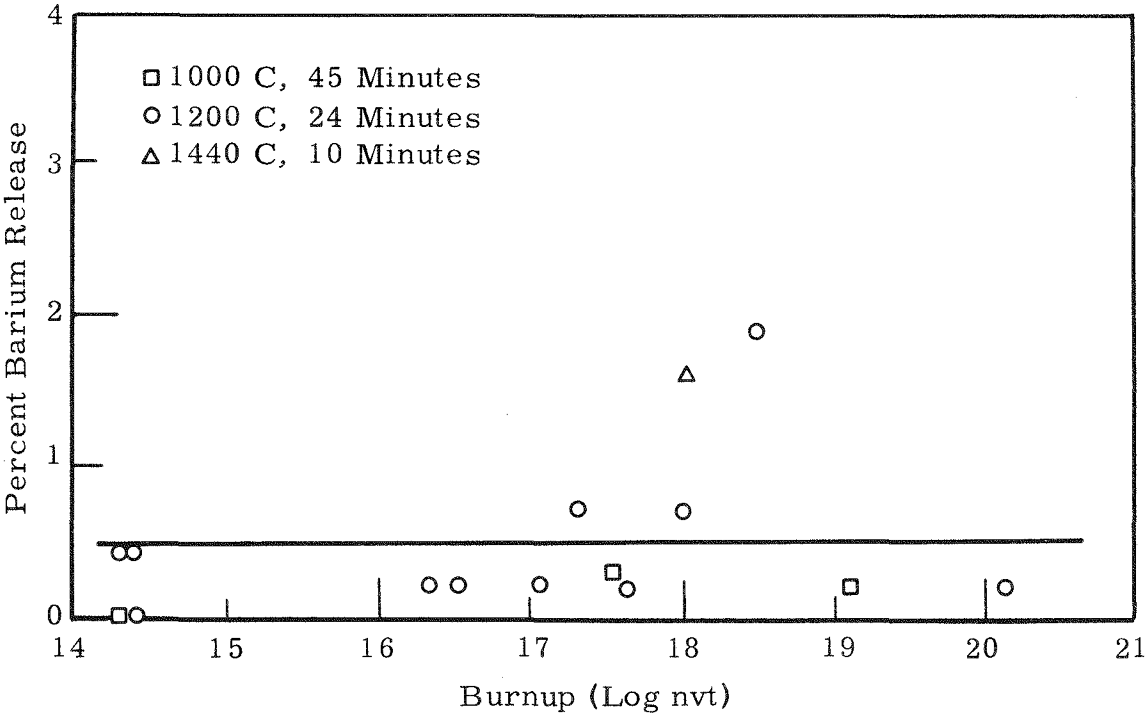

FIGURE 12

Release of Barium as a Function of Burnup

Appendix A shows that the strontium and barium results were quite scattered. This is probably due to analytical problems as indicated by strontium data of the last two tests and subsequent re-analysis of a few available samples of earlier tests by a revised method. Any slight trend caused by irradiation effects would be lost in the spread of the plotted data. As discussed later, burnup would not be expected to influence the release of strontium or barium.

Nonvolatile Elements

The elements of the "nonvolatile" group did not display a burnup effect. Figures 13 through 17 show the percentage release as a function of irradiation level for zirconium, cerium, neptunium, plutonium and uranium, respectively. The latter four nuclides were added to the analytical schedule for only the tests at higher irradiation levels to obtain the magnitude of their release. Consequently, the effect of burnup is not clearly displayed for these elements. However, from all considered aspects, irradiation level would not be expected to influence the fractional release materially for these nonvolatile elements.

Volatile Oxide Group

Two of the fission product elements investigated are unusual in that their oxides are much more volatile than the metals. Ruthenium boils at 4250 C; its tetraoxide boils at 135 C. Similarly, molybdenum boils at 4300 C while the sublimation point of molybdenum trioxide is 700 C.(14, 15) The extent to which these two fission products were released from heated uranium, therefore, depended upon the oxidation of the metal to the volatile oxide.

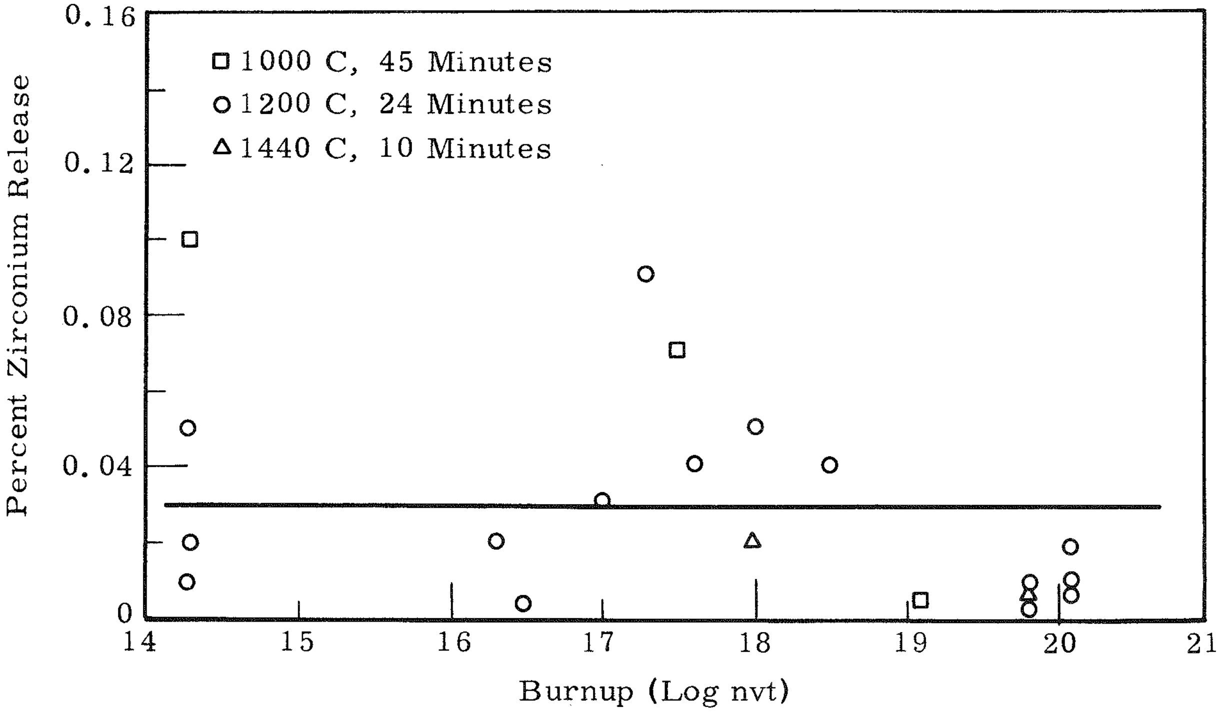

FIGURE 13

Release of Zirconium as a Function of Burnup

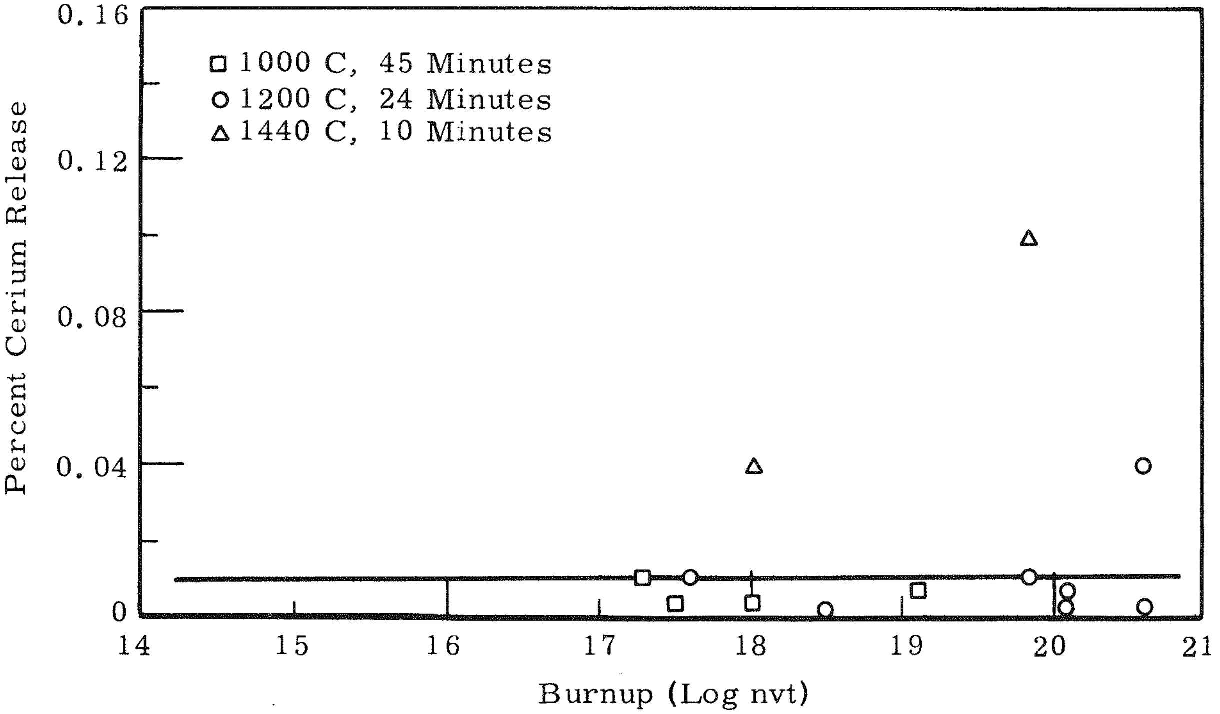

FIGURE 14

Release of Cerium as a Function of Burnup

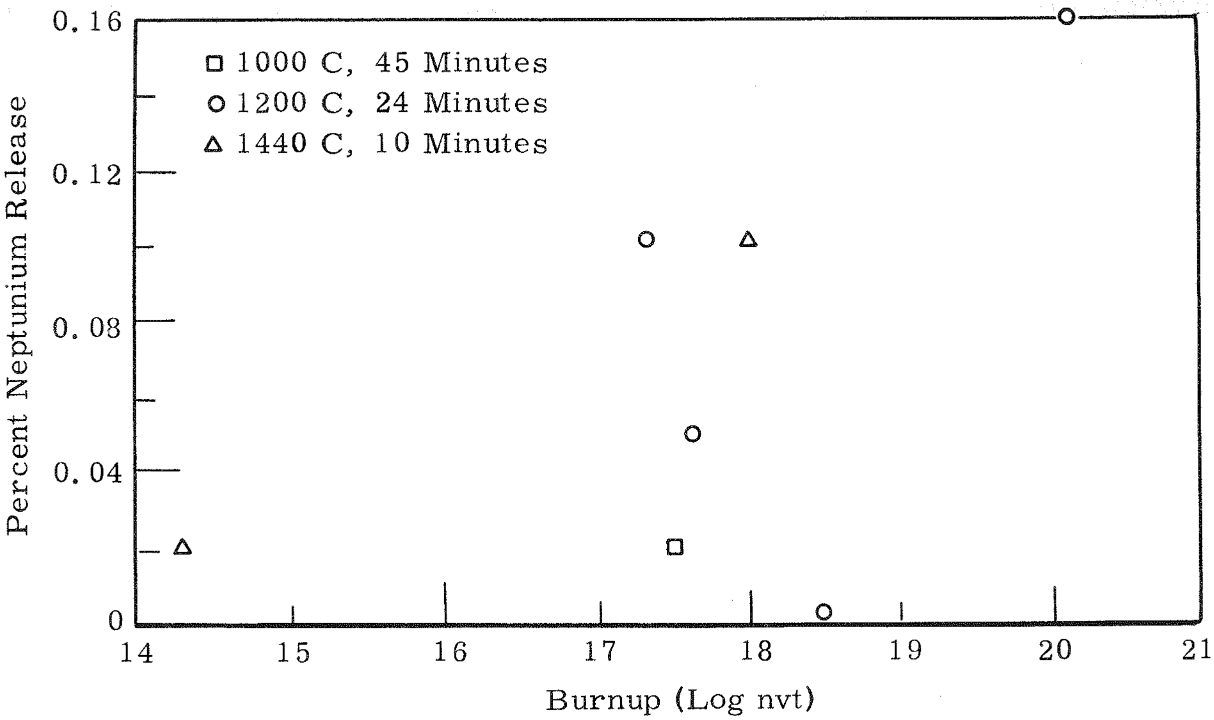

FIGURE 15

Release of Neptunium as a Function of Burnup

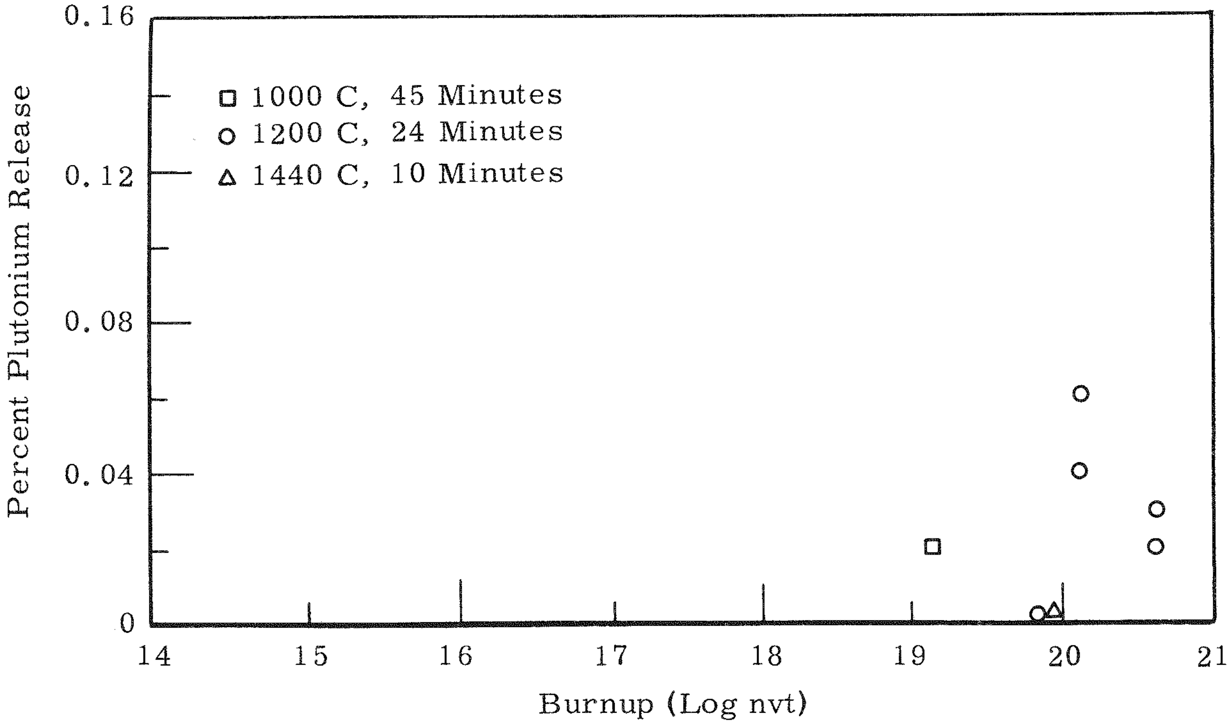

FIGURE 16

Release of Plutonium as a Function of Burnup

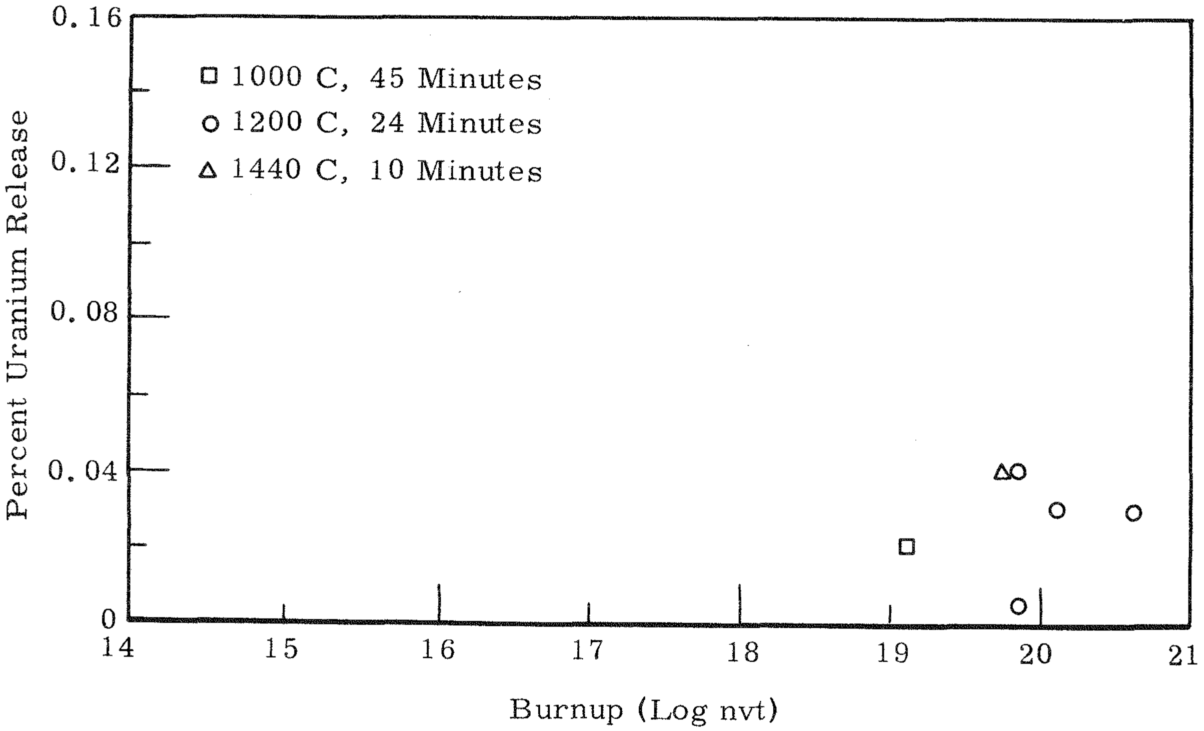

FIGURE 17

Release of Uranium as a Function of Burnup

Figures 18 and 19 present the ruthenium and molybdenum results, respectively. Molybdenum was not analyzed initially, so the data are fewer than for ruthenium. The percent releases were about 0.1 percent at irradiation levels up to 3×10^{18} nvt, above which the release increased until it was 5 percent at 4×10^{20} nvt. A possible explanation of these results is presented under Discussion.

Uranium Oxidation

The percent of the uranium specimen oxidized during each test was measured by chemical means and the results are reported in Appendix A.

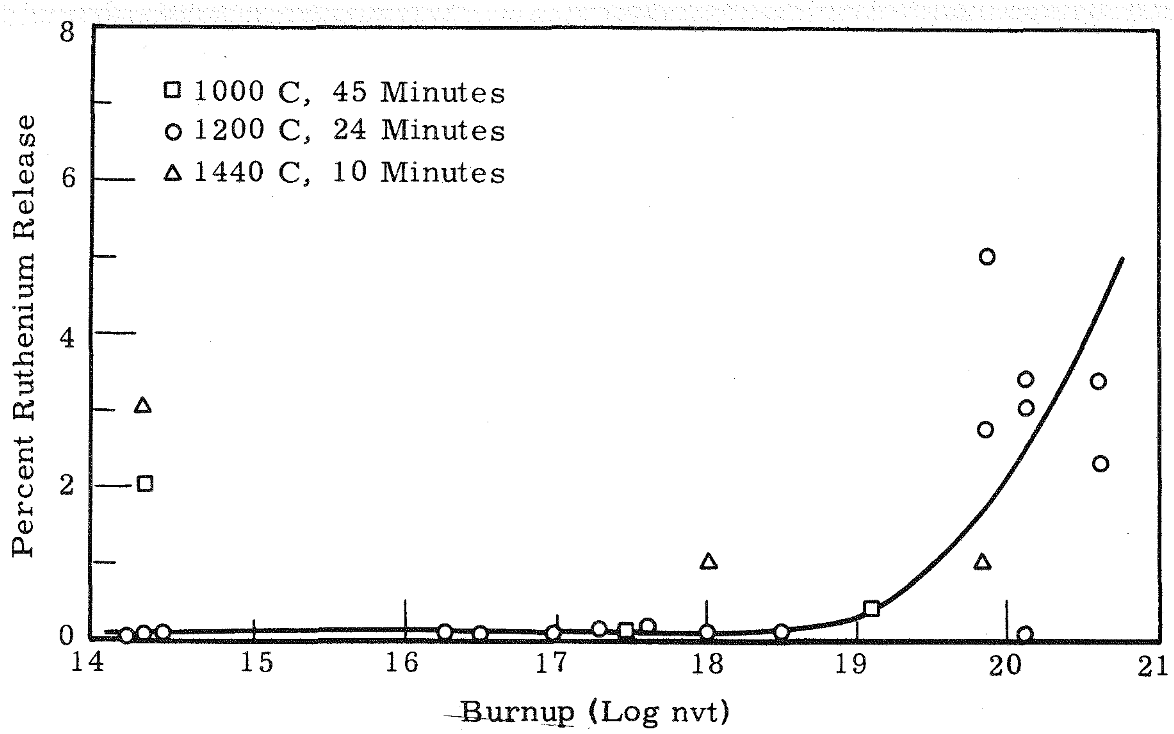

FIGURE 18

Release of Ruthenium as a Function of Burnup

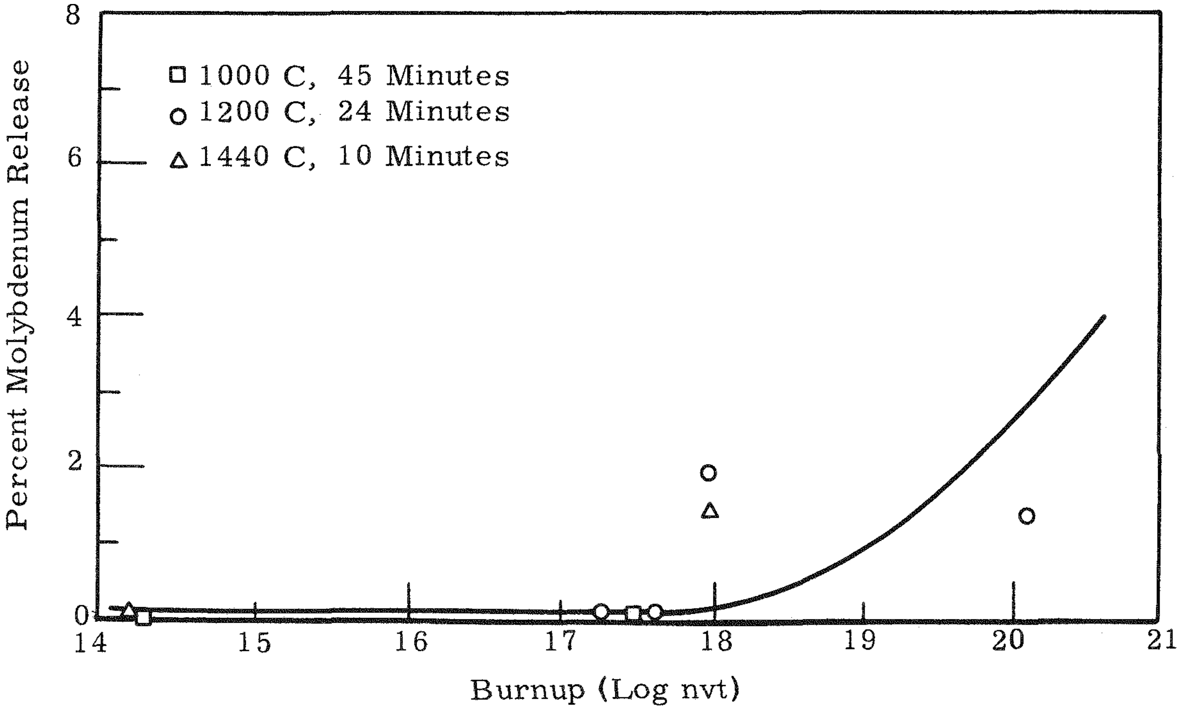

FIGURE 19

Release of Molybdenum as a Function of Burnup

Figure 20 shows graphically how the oxidation varied as a function of burnup. The oxidation rate remained constant at all irradiation levels up to about 10^{18} nvt; above this level the specimens were oxidized more extensively.

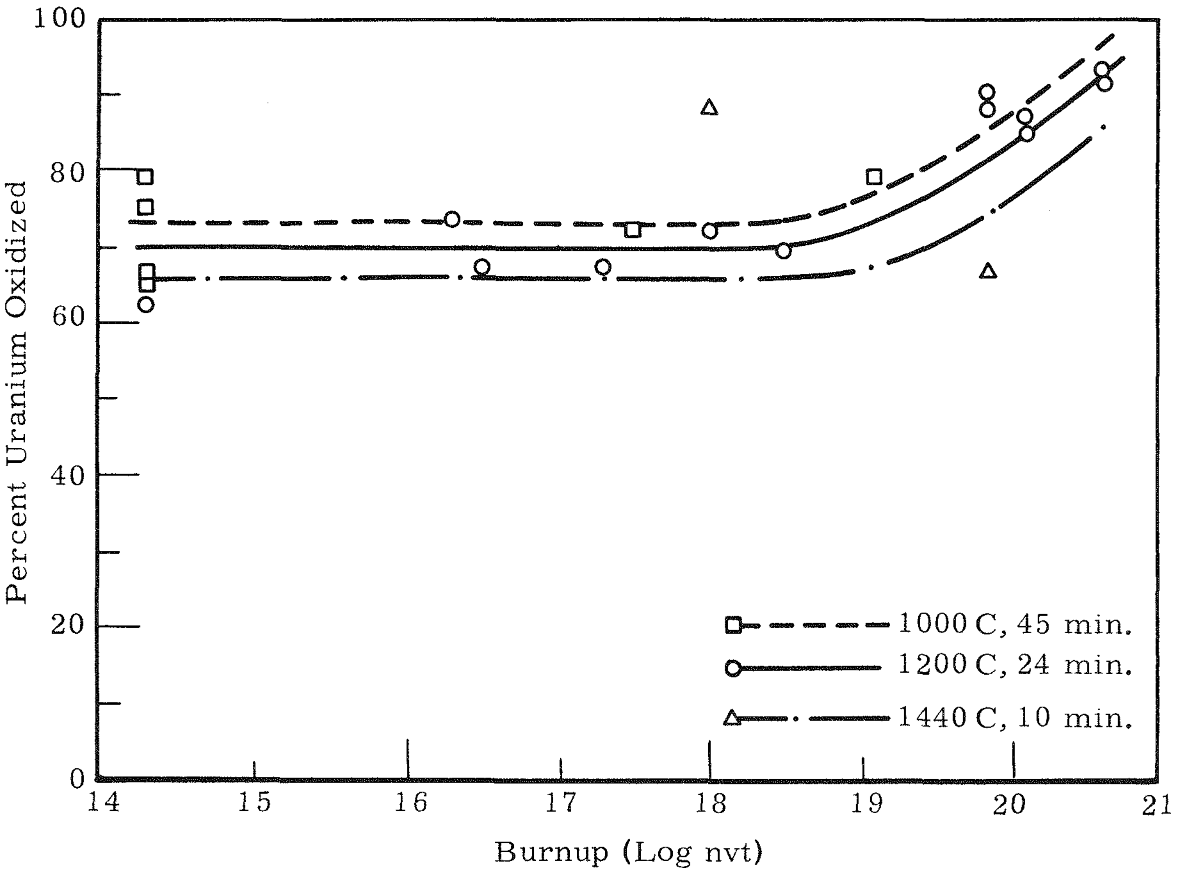

FIGURE 20

Effect of Burnup on Percent of Uranium Oxidized

Rate of Fission Product Release

Measurement of the fractional release of fission products from uranium as a function of time of heating was beyond the original scope of this study. The effect of time was shown in an earlier study on low level irradiated specimens to be such that the release was proportional to the uranium oxidation rate.(11, 13) This fact, coupled with the demonstrated influence of irradiation level on oxidation, suggested that the release rate of the volatile elements would be affected by time. Therefore, several of the tests were adapted to measure the releases of xenon and of cesium as a function of time.

Figure 21 is a graph of the charcoal trap counting rate as a function of time for three different irradiation levels. The graph shows that as the irradiation level increased, the charcoal trap attained its maximum counting rate earlier, until at the highest level of irradiation, the maximum was reached only three minutes after the uranium was melted. Since about one minute was required for gas travel time, the total release of xenon had occurred shortly after melting. These observations would be expected in view of the hypothesis of bursting fission gas bubbles dicussed in the following section.

Figure 22 shows the counting rate of a scintillation crystal placed next to the Millipore filter in Run AT-24. In this test the filter was placed between the furnace and the caustic scrubber. The instrument was biased to examine the energy region straddling that of cesium-137. The maximum counting rate was reached eight minutes after the specimen temperature reached the melting point. Unfortunately, similar techniques were not used during tests at lower irradiation levels, so a direct comparison to show the effect of irradiation level is not possible. However, in the early work of Hilliard(11) using uranium irradiated to 2×10^{14} nvt, the caustic pellet trap located at the furnace outlet was scanned during the runs by a Geiger-Mueller tube. The counting rate curve of such a test is plotted (Figure 22) for run A-11, which was an 85-minute test in air at 1215 C. It is evident that the radioactive content of the trap continued to build up for about 50 minutes, after which a constant level was maintained. Fifty minutes was the time required to completely oxidize the specimen at those conditions. Of course, the GM instrument indicated gamma events from all fission products present; however, iodine-131 and tellurium-132 were

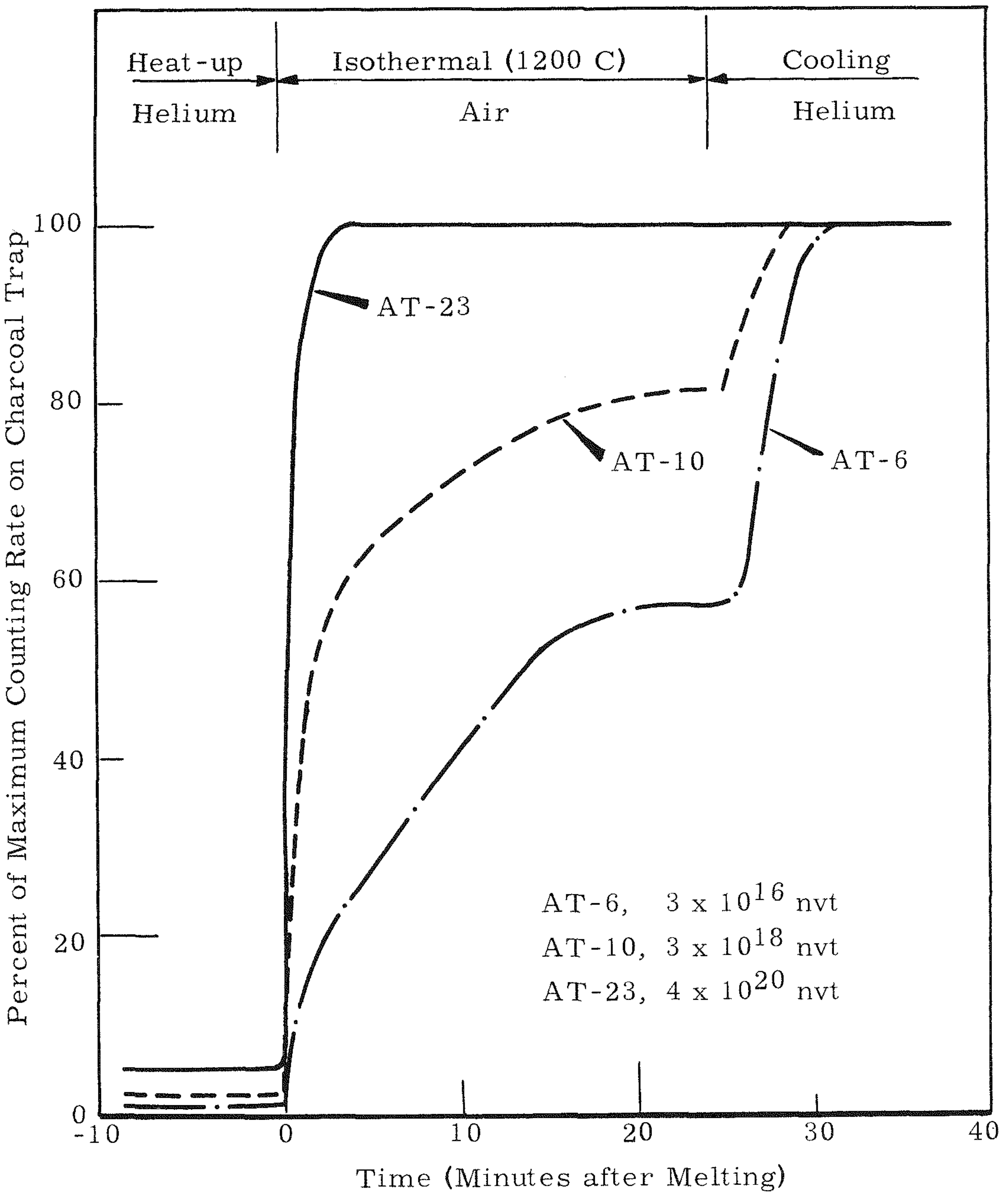

FIGURE 21

Effect of Burnup on the Xenon Release Rate

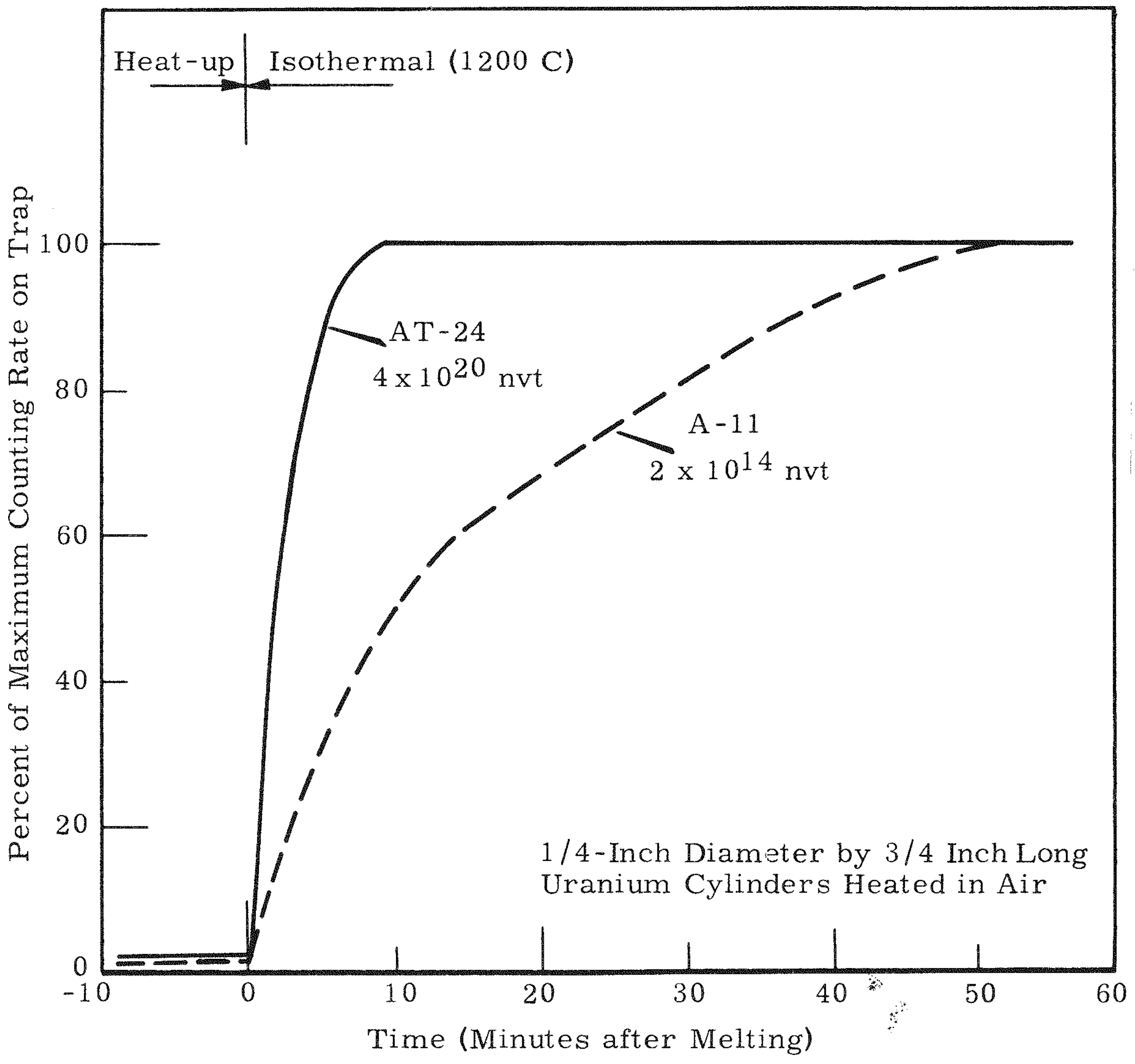

FIGURE 22

Effect of Burnup on Rate of Release of Volatile,
Nongaseous Elements

predominant. It is believed that the comparison shown in Figure 22 is valid; i.e., the fractional release of the "volatile" elements cesium, iodine and tellurium was much more rapid from highly irradiated uranium than from trace irradiated metal.

Deposition Pattern of Released Material

The radioactive material leaving the crucible was collected in a series of traps as shown in Figure 2. In some of the tests each trap was decontaminated separately and the washings analyzed individually. Thus, it was possible to obtain some cursory information concerning the deposition behavior of the released fission products. Although this information was incidental to the main objective and variables important to an intentional deposition study were not measured or controlled, some general trends were evident. Table II gives the percent of the released material which was collected on the furnace outlet leg, the two caustic scrubbers, the two cold traps, and the Millipore filter, in that order. The figures in the table are averages for eight tests where the specimen was heated to 1200 C.

TABLE II

DEPOSITION PATTERN OF RELEASED MATERIAL

	Percent of Released Material Collected			
Element	Furnace Outlet Leg	Caustic Scrubbers	Cold Traps	Millipore Filter
Iodine	20	77	2	1
Cesium	42	35	13	10
Tellurium	53	36	5	6
Strontium	25	40	15	20
Barium	30	35	20	15
Zirconium	35	25	25	15
Cerium	25	45	23	7
Neptunium	40	50	7	3
Ruthenium	26	70	2	2
Molybdenum	45	22	30	3

DISCUSSION

Irradiation conceivably could alter the uranium oxidation rate and the rate of escape of fission products from uranium in several ways. Creation of gas-filled cracks and bubbles, differences in fission product concentration, dimensional changes, and chemical reactions are possible causes for change in the release characteristics.

Fission Gas Bubble Formation

An important mechanism for accelerating the release of volatile fission products is the formation of a gaseous phase in the uranium matrix. This phenomenon is being studied extensively in relation to in-reactor swelling.(16, 17, 18) Although the nucleation step is not fully understood, it is known that once nucleated, fission gas bubbles behave as if their internal pressures were restrained by surface tension forces.(18) Bierlein and Mastel(17) photographed the bubbles in as-irradiated uranium at 0.2 percent burnup. They showed bubbles of 0.1 μ radius at a concentration of three bubbles per cubic micron. Pugh(16) reported that uranium irradiated to 0.1-0.45 percent burnup had 0.05 μ radius bubbles about 0.5 μ apart, or about eight per cubic micron. Equations are developed in Appendix B showing the relationship between bubble radius, bubble concentration and burnup. In Figure 23 the radius is plotted as a function of integrated neutron flux for two bubble concentrations. The data reported by Bierlein and by Pugh agree favorably with the theoretical values shown in Figure 23.

Solubility data(19) for xenon in uranium were used in conjunction with the hypothesis of equilibrium between the surface tension and gas pressure to derive an expression relating the percent of gas dissolved as a function of burnup. This relationship, plotted graphically in Figure 23, shows that 100 percent of the gas is dissolved at burnups less than 3×10^{15} nvt. Above this exposure bubbles nucleate at some concentration in the approximate range of 3 to 10 bubbles per cubic micron and grow rapidly in size to about 10^{-3} μ radius at 10^{16} nvt. Additional irradiation results in a continued, slower growth in bubble radius with a related decrease in the soluble gas

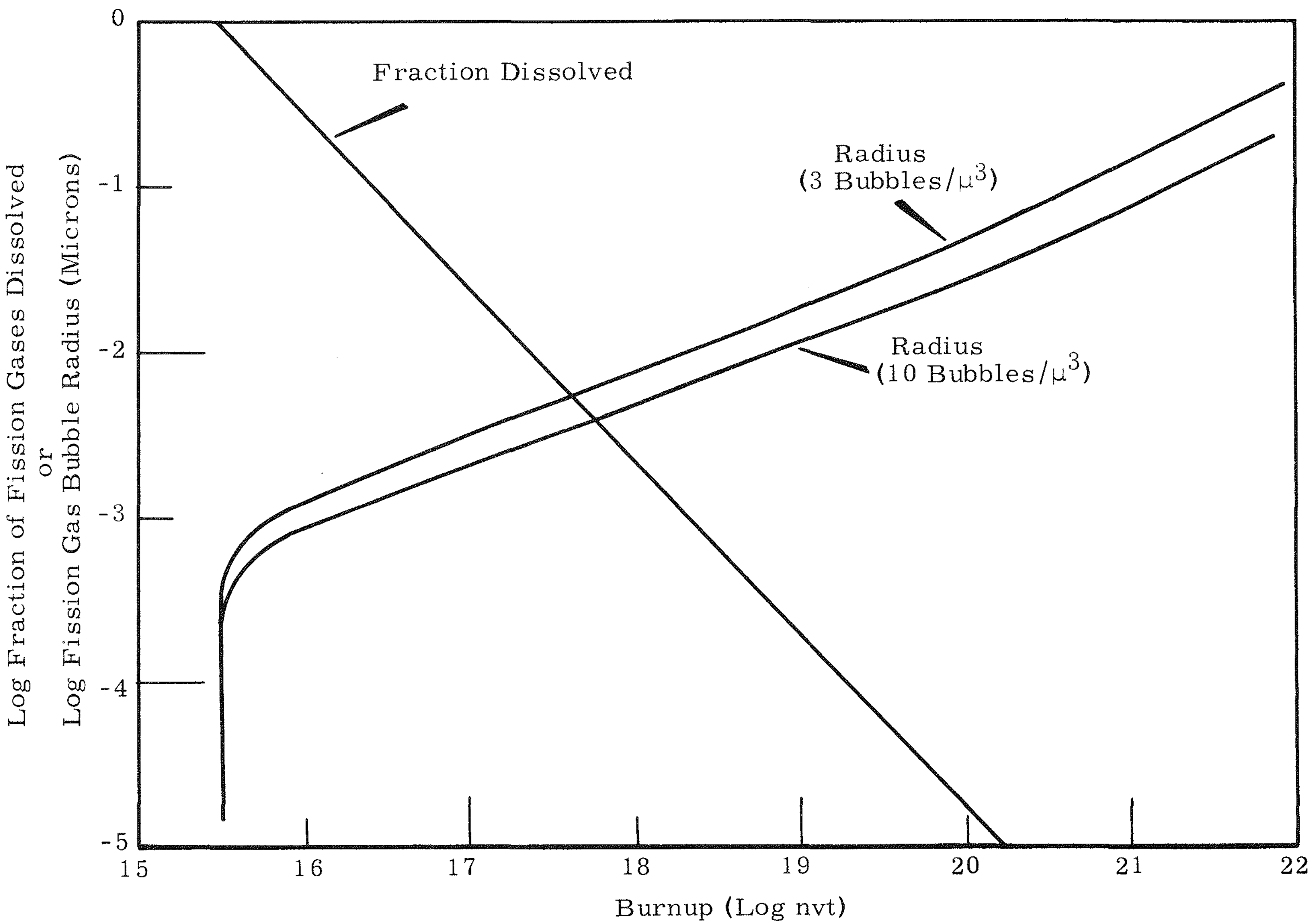

FIGURE 23

The Growth of Fission Gas Bubbles with Burnup fraction until at a burnup of 10^{21} nvt the radius is about 0.1 μ. At the highest burnup used in this study (4×10^{20} nvt) less than 0.001 percent of the fission gas was dissolved in the metal; essentially 100 percent was located in the bubbles.

The behavior of fission gas bubbles upon the melting of highly irradiated uranium was studied recently by Buddery and Scott.(20) They showed that the bubbles swelled, joined and burst through the surface, releasing 99.5^+ percent of the rare gas as the uranium melted. Metallographic studies of specimens quenched at various times prior to and following melting showed vividly the growth of the bubbles and subsequent escape of the fission gases.

Thus, both theoretical and experimental evidence confirm the importance of irradiation level on the release of volatile fission products through the formation of a two-phase (gas-metal) system.

Other Considerations

An obvious change in uranium caused by irradiation is the accumulation of fission products. The concentration of many fission product elements, including stable isotopes, is approximately proportional to the burnup. This is shown by Table III, which lists the theoretical concentrations at two irradiation levels--the lowest and the highest used in the experimental work. Activity coefficient data are lacking for the fission product-uranium systems, but ideality probably is approached closely at the very dilute concentrations existing in the test specimens. Indeed, McKenzie(21) showed that plutonium-uranium systems behaved ideally and the fractional volatilization of plutonium was independent of concentration. Concentration per se should not affect the fractional volatilization of fission products from uranium.

TABLE III

THEORETICAL CONCENTRATION OF FISSION PRODUCTS IN IRRADIATED URANIUM

Fission Product Element	Concentration (Atom Percent)	
	2×10^{14} nvt	4×10^{20} nvt
Xenon	1.9×10^{-8}	3.5×10^{-2}
Iodine	1.8×10^{-9}	1.9×10^{-3}
Cesium	1.4×10^{-8}	3.1×10^{-2}
Tellurium	2.0×10^{-9}	3.9×10^{-3}
Strontium	1.0×10^{-8}	1.5×10^{-2}
Barium	7.5×10^{-9}	9.3×10^{-3}
Zirconium	2.4×10^{-8}	5.4×10^{-2}
Cerium	1.1×10^{-8}	2.6×10^{-2}
Ruthenium	1.1×10^{-8}	1.5×10^{-2}
Molybdenum	1.5×10^{-8}	2.7×10^{-2}
Gross Fission Products	1.6×10^{-7}	3.2×10^{-1}

One mechanism for the escape of fission products from heated uranium is by diffusion to the surface and subsequent evaporation. The diffusion process is a combination of slow atomic diffusion through solid grains of metal and oxide and more rapid diffusion along intergranular cracks and pores. Irradiation possibly increases the diffusion rate by (1) creating vacancies within the grains and thus enhancing atomic diffusion, (2) increasing the proportion of fission products located interstitially, and (3) increasing the number and size of the cracks and pores. This increase in diffusion rate will cause an increase in the fractional release only if diffusion is the rate-limiting step.

Macro changes in the uranium during irradiation, such as swelling, growth and surface roughening, could alter the fission product release. While some surface pitting was observed with the more highly irradiated specimens, the related effect was short-lived and insignificant due to rapid surface oxidation under the conditions of test. Dimensional changes by anisotropic growth were observed to be minor. Swelling from the production of solid fission products would have been <2 percent by volume(16) or about a 1 percent increase in the surface area--a negligible change. Consequently, any effect on the release of fission products from dimensional changes in the specimens would be lost in the variability of measurements.

Formation of stable compounds, e. g., CsI or UI_4, could change the volatility of the combined elements. Very little is known about the chemical nature of fission product elements produced during irradiation, but the extremely low concentrations (Table III) preclude combinations in amounts that would significantly alter their release.

Retention of fission products *in situ* by absorption in the uranium oxide is another possible cause for the observed burnup effect. In earlier work(11) with specimens having low burnup, the completely oxidized uranium retained 20-30 percent of the xenon, iodine and tellurium. This suggested that xenon was trapped as a clathrate and iodine and tellurium were absorbed in the oxide. If the amount retained in the uranium oxide increased at a lesser rate than the increase in total atoms produced by higher irradiations, then an increase in the fractional release would result.

Explanation of Results

The experimental results reported in Appendix A and in Figures 7 through 22 can be interpreted in terms of the preceding discussions.

Uranium Oxidation

The apparent influence of irradiation level on the uranium oxidation rate, as shown in Figure 20, is explained by the formation and growth of bubbles. Uranium specimens were heated to temperature in flowing impure helium with air admitted to the system just prior to reaching the uranium melting point. This produced a thin layer of oxide. With lightly irradiated uranium specimens (up to 5×10^{18} nvt), the continuity and strength of this oxide layer was sufficient to retain the molten metal. Since the original cylindrical shape was approximately maintained throughout the test, the surface area was relatively uniform between tests and the percent oxidized was relatively constant. With highly irradiated specimens ($\sim 10^{20}$ nvt) the initially larger bubbles and subsequent growth with heating burst the oxide barrier. This allowed the molten uranium to flow and cover the bottom of the crucible. The increase in surface area, by an approximate factor of 1.3, produced a higher total oxidation for the same time and temperature conditions. Bubbles too small to cause the flow of uranium could crack and distort the oxide layer sufficiently to alter the diffusion path of the oxygen and result in an increased oxidation rate. The irradiation level at which a significant increase in the oxidation would be produced by gas bubble growth was not predictable from the data, but apparently was between 5×10^{18} and 1×10^{20} nvt. This observed relationship of oxidation and irradiation level would not be expected to be as pronounced with large bare uranium pieces where the strength of the oxide layer would not be sufficient to prevent the free flow of uranium regardless of irradiation level.

Volatile Oxide Group

The significant increase in the release of ruthenium and molybdenum at irradiation levels above 5×10^{18} nvt (Figures 18, 19) can be explained by the similar increase in uranium oxidation (Figure 20). Tests with trace-irradiated specimens(4, 11) showed that after all the uranium had oxidized ruthenium release accelerated greatly. Apparently, RuO_4 is not formed in the presence of uranium metal or lower uranium oxides. The increased surface area to volume ratio caused by bursting fission gas bubbles and the flowing of the uranium provided a mechanism for hastening the oxidation of ruthenium and molybdenum to their volatile oxides. Therefore, the sharp increase in the release shown in Figure 18 is not directly caused by irradiation but is a function of the kinetics of the ruthenium oxidation reaction.

Volatile Elements

The greatest effect of fission gas bubbles on fission product release was on the volatile elements group--the rare gases, the halogens and the alkali metals. This is to be expected since the major portion of these elements is in the gas bubbles. Figure 23 shows that a negligible amount of the xenon was dissolved in the metal for irradiations above 10^{18} nvt. Solubility data for iodine and cesium in uranium are not available, but it is reasonable to assume that at high burnup and an irradiation temperature of $\sim$400 C these elements exist largely in the vapor space in the bubbles. Thus, these materials were released rapidly as the bubbles expanded and reached the surface. Both the increase in fractional release shown in Figures 7, 8 and 9 and the increase in the fractional release rate indicated in Figures 21 and 22 can be explained by the existence of gas bubbles in the irradiated uranium.

The release of volatile elements was shown to be dependent on irradiation level even at burnups too low for the fission gas bubbles to have much effect. Absorption in the uranium oxide and increased diffusivity must combine with the fission gas bubbles to give the observed over-all effect.

Tellurium release would not be influenced by the presence of bubbles because, being relatively nonvolatile in the elemental form, tellurium would not concentrate in the bubbles to any significant extent.

Nonvolatile and Slightly Volatile Elements

The release of the elements in the "nonvolatile" and "slightly volatile" groups should not be significantly affected by fission gas bubble formation. Their release rate is probably controlled by an evaporation step rather than by diffusion. Oxidation was only increased by a factor of 1.3, and a similar increase in release of these elements would not be noticed because of experimental variations.

CONCLUSIONS

The influence of irradiation level on the fractional release of fission products from metallic uranium must be considered minor when compared to other variables, such as temperature, time and atmosphere. However, the release of the volatile fission product elements as a function of time is much greater from highly irradiated metal, with almost quantitative escape of the fission gases at the time of fuel melting.

Although the fractional release of all the fission product elements is not markedly affected by burnup, the trend for higher releases of certain elements with higher irradiation levels should be considered when applying experimental data obtained from trace-irradiated specimens.

The chief reason for a burnup effect is the formation of bubbles of fission gases. These bubbles contain essentially all the rare gases and halogens and a large fraction of the cesium. Their size becomes sufficiently large at irradiation levels above 10^{18} nvt to change the major escape process from atomic diffusion to migration through the expanded bubbles and cracks and degasification.

The uranium oxidation rate may be increased by high burnup because of an increase in surface area to weight ratio caused by the expansion of the fission gas bubbles.

Oxide and metal alloy fuels probably are subject to a burnup effect similar to that determined in this study for metallic uranium. The magnitude of such an effect is impossible to predict.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance in planning and performing the experiments received from C. E. Linderoth, H. G. Rieck and A. J. Scott. Specimen irradiations were overseen by R. K. Marshall and sample analyses were performed by the staff of Radiochemical Analysis Operation under the special guidance of A. C. Leaf.

APPENDIX A

TABULATION OF ANALYTICAL RESULTS

Run No.	nvt	Percent Oxidized	Percent Released from Specimen												
			Xe	I	Cs	Te	Sr	Ba	Ru	Mo	Zr	Ce	Np	Pu	U
Temperature = 1000 C, Time = 45 min.															
AT-15	2×10^{14}	75	82	15	30	58	1.7	0.01	2.0	0.02	0.1				
AT-11	3×10^{17}	72	94	78	50	50	0.07	0.3	0.1	0.02	0.07	0.004	0.02		
AT-19	1.2×10^{19}	79	99	84	55	44	0.1	0.2	0.4		0.005	0.006		0.015	0.016
Temperature = 1200 C, Time = 24 min.															
AT-2	2×10^{14}	79	84	70	37	70	0.4	0.4	0.1		0.02				
AT-3	2×10^{14}	67	68	38	20	70	0.03	0.06	0.07		0.01				
AT-4	2×10^{14}	62	73	74	34	66	0.3	0.4	0.1		0.05				
AT-5	2×10^{16}	73	91	80	45	72	0.1	0.2	0.08		0.02				
AT-6	3×10^{16}	67	92	75	44	74	0.2	0.2	0.07		0.004				
AT-7	1×10^{17}		95	85	40	65	0.2	0.2	0.05		0.03				
AT-13	2×10^{17}	67	95	65	57	75	0.6	0.7	0.1	0.03	0.09	0.01	0.1		
AT-8	4×10^{17}		95	85	35	50	0.1	0.2	0.2	0.08	0.04	0.01	0.05		
AT-9	1×10^{18}	72	98	75	68	45	0.4	0.7	0.04	1.9	0.05	0.003	0.6		
AT-10	3×10^{18}	69	97	93	55	60	1.2	1.9	0.06		0.04	0.001	0.003		
AT-20	7×10^{19}	88	99.8	97	90	62	1.3	*	2.8		0.002	0.2		0.003	0.04
AT-22	7×10^{19}	90		*	94	50	0.2	*	5.0		0.01	0.01			0.005
AT-18	1×10^{20}	87	99.5	99.5	68	84	0.07	0.2	3.4		0.02	0.002		0.057	0.03
AT-16	1.4×10^{20}	*	99.8	94	75	54	*	*	3.0	1.4	0.008	*	0.2		
AT-17	1.4×10^{20}	85	99.6	96	87	75	*	*	0.07		0.01	0.006	2.0	0.04	
AT-23	4×10^{20}	91			75	21	0.2		2.3			0.002		0.02	0.03
AT-24	4×10^{20}	92			80	24	0.3		3.4			0.04		0.03	0.27
Temperature = 1440 C, Time = 10 min.															
AT-14	2×10^{14}	66	70	70	40	20	1.1	*	*	0.1	*	*	0.2		
AT-12	1×10^{18}	88	98	77	53	16	2.4	1.6	1.0	1.4	0.02	0.04	0.1		
AT-21	7×10^{19}	66	99.8	98	98	27	*	*	1.0		0.004	0.1		0.003	0.04

*Anomalous data not reported

APPENDIX B

FISSION GAS BUBBLE FORMATION

Bases for Calculations

(1) The term "fission gas" is defined as all of the rare gases and halogens.

(2) Irradiation temperature of 400 C.

(3) Equilibrium always exists between the internal gas pressure and the surface tension forces surrounding a bubble.(18) Surface tension of uranium is $\sim 10^3$ ergs/cm^2.

(4) Bubbles are of uniform size.(16, 17)

(5) Bubbles nucleate in an approximate concentration range of 3 to 10 per cubic micron(16, 17) and remain at this concentration.

(6) Uniform neutron flux distribution, resulting in homogeneous mixture of fission gas atoms and uranium.

(7) The Ostwald coefficient, k, equals 10^{-8},(19), where $k = \frac{x}{y}$

x = number of fission gas atoms dissolved in 1 cc uranium,

y = number of fission gas atoms per cc of bubble.

(8) The fission gas generation rate is proportional to burnup (Table III, text).

These assumptions are reasonably valid, with numbers 4, 5 and 7 being questionable.

The pressure within each gas bubble is determined by the equation of state:

$$p = \frac{CnRT}{V_i} \tag{1}$$

where

C = "compressibility factor" obtained from reduced pressure and temperature data for xenon.(22)

p = pressure, atm

n = moles of fission gas per bubble = N_i/A

N_i = number of fission gas atoms per bubble

A = Avogadro's number = 6×10^{23} atoms/mole

R = gas constant = 82 cc-atmospheres $\text{mole}^{-1} K^{-1}$

T = temperature, K

V_i = volume of gas bubble, cc.

$$V_i = \frac{4}{3} \pi r^3 = 4.2\ r^3 \tag{2}$$

where

r = bubble radius, cm.

Substituting (2) into (1)

$$p = \frac{CN_iRT}{4.2\ A\ r^3} \tag{1a}$$

Assuming that equilibrium exists between gas pressure and surface tension forces,

$$p = \frac{2\gamma}{r} \tag{3}$$

where

γ = surface tension = 10^3 erg/cm^2 = 10^{-3} cc-atm/cm^2

$$p = \frac{2 \times 10^{-3}}{r} \text{ atm} \tag{3a}$$

combining (1a) and (3a)

$$r^2 = \frac{C\ N_i\ R\ T}{8.4 \times 10^{-3}\ A} \tag{4}$$

By definition,

$$y = \frac{\text{number fission gas atoms}}{\text{cc of bubble}}$$

$$y = \frac{N_i}{V_i} = \frac{N_i}{4.2\ r^3} \tag{5}$$

combining (4) and (5)

$$y = \frac{2.17 \times 10^{16}}{C\ r} \tag{6}$$

From Ostwald coefficient,

$$x = 10^{-8} y$$

$$x = \frac{2.17 \times 10^{8}}{C\ r} \qquad (7)$$

The compressibility factor, C, is a function of reduced temperature and pressure as defined in Reference 22.

$$C = 1, \qquad p < 150 \text{ atm} \qquad (8)$$

$$C = 7.32 \times 10^{-4} p + 0.775, \qquad p > 150 \text{ atm} \qquad (8a)$$

or

$$C = \frac{1.464 \times 10^{-6}}{r} + 0.775, \qquad p > 150 \text{ atm} \qquad (8b)$$

Therefore, for pressures less than 150 atmospheres, Equation (6) becomes

$$y = \frac{2.17 \times 10^{16}}{r}, \qquad p < 150 \text{ atm} \qquad (9)$$

and for pressures greater than 150 atmospheres,

$$y = \frac{2.17 \times 10^{16}}{1.46 \times 10^{-6} + 0.775\ r}, \qquad p > 150 \text{ atm} \qquad (9a)$$

Similarly,

$$x = \frac{2.17 \times 10^{8}}{r}, \qquad p < 150 \text{ atm} \qquad (10)$$

$$x = \frac{2.17 \times 10^{8}}{1.46 \times 10^{-6} + 0.775\ r}, \qquad p > 150 \text{ atm} \qquad (10a)$$

Bubble Radius as a Function of Burnup

A total fission gas atom balance gives

Total Atoms = Atoms in bubbles + Atoms dissolved in metal (11)

$$N_o = V_b y + V_m x$$

where

N_o = total fission gas atoms per cc of uranium
V_b = total gas volume per cc of metal, cc/cc
V_m = uranium volume, taken as 1 cc/cc of gas plus metal

From Table III, text,

$$N_o = 5 \times 10^{-2} \text{ (nvt)} \tag{12}$$

and,

$$V_b = N_b V_i = 4.2 N_b r^3 \tag{13}$$

where

N_b = number of bubbles per cm^3.

Combining (11), (12) and (13)

$$N_o = 5 \times 10^{-2} \text{ (nvt)} = 4.2 N_b r^3 y + x \tag{14}$$

which reduces to

$$\text{nvt} = (8.4 \times 10^9 N_b r^3 + 20) x. \tag{15}$$

Substituting Equations (10) and (10a) into (15),

$$\text{nvt} = 1.82 \times 10^{18} N_b r^2 + \frac{4.34 \times 10^9}{r}, \quad p < 150 \text{ atm} \tag{16}$$

and

$$\text{nvt} = \frac{1.82 \times 10^{18} N_b r^3 + 4.34 \times 10^9}{1.46 \times 10^{-6} + 0.775 r}, \quad p > 150 \text{ atm} \tag{16a}$$

Equations (16) and (16a) give the relationship between bubble radius, bubble concentration and burnup. Although they cannot be solved explicitly for the radius as a function of nvt, they can be plotted graphically with bubble concentration, N_b, as the parameter. Figure 23 in the text is such a plot.

Fraction of Gas Dissolved

$$F = \frac{x}{N_o} \tag{17}$$

where

F = fraction of gas dissolved in uranium.

Combining with Equations (10), (10a) and (12)

$$F = \frac{4.34 \times 10^9}{(nvt)(r)}, \qquad p < 150 \text{ atm} \qquad (18)$$

and

$$F = \frac{4.34 \times 10^9}{(nvt)(1.46 \times 10^{-6} + 0.775\, r)}, \qquad p > 150 \text{ atm} \qquad (18a)$$

Using the relationship between r and nvt expressed in Equations (16) and (16a), and by Figure 23, the fraction dissolved was plotted as a function of (nvt) in Figure 23. Notice that the fraction dissolved is independent of bubble concentration.

Equations (18) and (18a) reduce approximately to

$$F = \frac{2.97 \times 10^{15}}{(nvt)}, \qquad nvt > 3 \times 10^{15} \qquad (19)$$

REFERENCES

(1) Parker, H. M. and J. W. Healy. "Environmental Effects of a Major Reactor Disaster", *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy*, Geneva, 13: 106-109. New York: U. N. Pub., 1955.

(2) USAEC. *Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants*, WASH-740. March, 1957.

(3) Creek, G. E., et al. *Experiments on the Release of Fission Products from Molten Reactor Fuels*, ORNL-2616. July 22, 1959.

(4) Parker, G. W., et al. *Fuel Element Catastrophe Studies: Hazards of Fission Product Release from Irradiated Uranium*, CF-60-6-24. August 31, 1960.

(5) Parker, G. W., et al. *Fission Product Release from UO_2 by High Temperature Diffusion and Melting in Helium and Air*, CF-60-12-14. February 14, 1961.

(6) Rodgers, S. J. and G. E. Kennedy. *Fission Product Release During a Simulated Meltdown of a PWR Type Core*, NP-7071. October 20, 1958.

(7) Conn, P. K. *Summary of Fission Product Release Data from GE-ANPD Fuel Element Reactor Tests*, APEX-544. February, 1959. (Secret)

(8) Megaw, W. J., et al. "The Oxidation and Release of Iodine-131 from Uranium Slugs Oxidizing in Air and Carbon Dioxide", Reactor Science and Technology, *J. of Nuc. Energy, Parts A/B*, Vol. 15: 176-184. 1961.

(9) U. S. Air Force Nuclear Aircraft Research Facility. *Fission Products Field Release Test--I*, AFSWC-TR-59-44. September, 1959.

(10) U. S. Air Force Nuclear Aircraft Research Facility. *Fission Products Release Test--II*, AFSWC-TR-60-26. September, 1960.

(11) Hilliard, R. K. *Fission Product Release from Uranium Heated in Air*, HW-60689. August 3, 1959.

(12) Scott, A. J. *Fission Product Release by the High Temperature Uranium Steam Reaction*, HW-62604. November 18, 1959.

(13) Hilliard, R. K., et al. "Fission Product Release from Overheated Uranium--A Laboratory Study", *Health Physics* Vol. 7: 1-10. December, 1961.

(14) Glassner, A. The Thermochemical Properties of the Oxides, Fluorides and Chlorides to 2500° K, ANL-5750. 1957.

(15) Loftness, R. L. A Vapor Pressure Chart for Metals, NAA-SR-132. June 1, 1951.

(16) Pugh, S. F. "Swelling in Alpha Uranium Due to Irradiation", J. of Nuclear Materials, Vol. 4, No. 2: 177-199. July, 1961.

(17) Bierlein, T. K. and B. Mastel. Microstructural Changes in Irradiated Uranium Dioxide, HW-SA-1717. September 23, 1959.

(18) Greenwood, G. W., et al. "The Role of Vacancies and Dislocations in the Nucleation and Growth of Gas Bubbles in Irradiated Fissile Material", J. of Nuclear Materials, Vol. 1, No. 4: 305-324. December, 1959.

(19) Johnson, G. W. and R. Shuttleworth. "The Solubility of Krypton in Liquid Lead, Tin and Silver", Philosophical Magazine, Vol. 4: 957-963. August, 1959.

(20) Buddery, J. H. and K. T. Scott. "A Study of the Melting of Irradiated Uranium", J. of Nuclear Materials, Vol. 5, No. 1: 81-93. January, 1962.

(21) McKenzie, D. E. "The Volatilization of Plutonium from Neutron-Irradiated Uranium", Canadian J. of Chemistry, Vol. 34: 515-522. April, 1956.

(22) Dodge, B. F. Chemical Engineering Thermodynamics, McGraw-Hill Book Co., Inc: 159-162. New York, 1944.

INTERNAL DISTRIBUTION

Copy Number	
1	G. J. Alkire
2	T. K. Bierlein
3	J. H. Brown
4	E. W. Christopherson
5	L. F. Coleman
6	E. A. Evans - W. E. Roake
7	R. F. Foster
8	C. C. Gamertsfelder
9	O. H. Greager
10	W. A. Haney
11	H. Harty
12	O. F. Hill
13	R. K. Hilliard
14	E. R. Irish
15	G. Jansen, Jr.
16	R. L. Junkins
17	A. R. Keene
18	G. A. Last
19	M. C. Leverett
20	C. E. Linderoth
21	R. L. Moore
22	E. G. Pierick
23	W. H. Reas
24	D. L. Reid
25	L. C. Schwendiman
26	J. M. Skarpelos
27	W. H. Swift
28	J. C. Tobin
29	R. E. Trumble
30	F. W. Van Wormer
31	M. T. Walling, Jr.
32	E. C. Watson
33	L. L. Zahn
34	Technical Publications
35	300 Files
36	Record Center
37 - 61	Extra

EXTERNAL DISTRIBUTION (SPECIAL)

Number of Copies	
1	Argonne National Laboratory 9700 South Cass Avenue Argonne, Illinois Attn: R. C. Vogel
3	AEC-HOO Attn: P. G. Holstead M. J. Schneller Technical Information Library
2	Atomic Energy Commission Washington Attn: W. G. Fisher J. A. Lieberman
2	G. E. Technical Data Center, Schenectady
1	J. W. Healy - Consultant, Technical Hazards 570 Lexington, Room 2207 New York 22, N. Y.
3	Union Carbide Nuclear Company (ORNL) P. O. Box X Oak Ridge, Tennessee Attn: C. J. Barton W. E. Browning G. W. Parker
1	G. E. Vallecitos Attn: L. P. Bupp

EXTERNAL DISTRIBUTION

Number of Copies	
12	Aberdeen Proving Ground
1	Aerojet-General Corporation
1	Aerojet-General Nucleonics
6	Aeronautical Systems Division
2	Air Force Special Weapons Center
1	Alco Products, Inc.
1	Allis-Chalmers Manufacturing Company
1	Allis-Chalmers Manufacturing Company, Washington
1	Allison Division--GMC
4	Argonne Cancer Research Hospital
10	Argonne National Laboratory
4	Army Chemical Center
1	Army Chemical Center (Taras)
1	Army Chemical Corps
1	Army Environmental Hygiene Agency
1	Army Medical Research Laboratory
1	Army Signal Research and Development Laboratory
1	Atomic Bomb Casualty Commission
1	AEC Scientific Representative, France
1	AEC Scientific Representative, Japan
3	Atomic Energy Commission, Washington
4	Atomic Energy of Canada Limited
4	Atomics International
2	Babcock and Wilcox Company
2	Battelle Memorial Institute
1	Beryllium Corporation
2	Brooke Army Medical Center
4	Brookhaven National Laboratory
1	Bureau of Medicine and Surgery
1	Bureau of Mines, Albany
1	Bureau of Mines, Salt Lake City
1	Bureau of Ships (Code 1500)
1	Bureau of Yards and Docks
1	Chicago Patent Group
1	Columbia University (Rossi)
1	Combustion Engineering, Inc.
1	Combustion Engineering, Inc. (NRD)
1	Committee on the Effects of Atomic Radiation
2	Convair Division, Fort Worth
3	Defence Research Member
1	Defense Atomic Support Agency, Washington
1	Division of Raw Materials, Washington
1	Dow Chemical Company (Rocky Flats)
3	duPont Company, Aiken

TID-4500, 17th Ed.
UC-41, Health and Safety

EXTERNAL DISTRIBUTION (Contd.)

Number of Copies	
1	duPont Company, Wilmington
1	Edgerton, Germeshausen and Grier, Inc., Goleta
1	Edgerton, Germeshausen and Grier, Inc., Las Vegas
1	Frankford Arsenal
1	Franklin Institute of Pennsylvania
1	General Atomic Division
2	General Electric Company (ANPD)
1	General Electric Company, St. Petersburg
1	General Telephone and Electronic Laboratories, Inc.
1	Gibbs and Cox, Inc.
1	Glasstone, Samuel
1	Goodyear Aircraft, Akron (BUWEPS)
1	Goodyear Atomic Corporation
1	Grand Junction Office
1	Hawaii Marine Laboratory
1	Hughes Aircraft Company
1	Iowa State University
1	Journal of Nuclear Medicine
1	Kelly Air Force Base
3	Knolls Atomic Power Laboratory
1	Lockheed Aircraft Corporation
2	Los Alamos Scientific Laboratory
1	Lovelace Foundation
1	Lowry Air Force Base
1	M & C Nuclear, Inc.
1	Mallinckrodt Chemical Works
1	Maritime Administration
1	Martin Company
1	Massachusetts Institute of Technology (Hardy)
1	Mound Laboratory
1	National Academy of Sciences
1	NASA Lewis Research Center
2	NASA Scientific and Technical Information Facility
2	National Bureau of Standards
1	National Cancer Institute
1	National Distillers and Chemical Corporation
1	National Distillers and Chemical Corporation, Ashtabula
1	National Lead Company of Ohio
1	National Library of Medicine
1	Naval Hospital
1	Naval Medical Research Institute
1	Naval Ordnance Laboratory
1	Naval Postgraduate School
2	Naval Radiological Defense Laboratory
3	Naval Research Laboratory
1	New Brunswick Area Office
1	New York Operations Office

EXTERNAL DISTRIBUTION (Contd.)

Number of Copies	
1	New York University (Eisenbud)
1	Nuclear Materials and Equipment Corporation
1	Oak Ridge Institute of Nuclear Studies
15	Office of Naval Research
1	Office of Naval Research (Code 422)
1	Office of the Chief of Naval Operations
1	Office of the Surgeon General
1	Ordnance Tank-Automotive Command
1	Patent Branch, Washington
6	Phillips Petroleum Company (NRTS)
1	Power Reactor Development Company
3	Pratt and Whitney Aircraft Division
1	Princeton University (White)
2	Public Health Service
1	Public Health Service, Las Vegas
1	Public Health Service, Montgomery
1	Quartermaster Food and Container Institute
1	Quartermaster Research and Engineering Command
1	States Marine Lines, Inc.
1	Strategic Air Command
1	RAND Corporation
1	Rensselaer Polytechnic Institute
1	Research Analysis Corporation
1	Rocky Mountain Arsenal
1	Sandia Corporation, Albuquerque
1	Sandia Corporation, Livermore
1	Schenectady Naval Reactors Operations Office
1	Second Air Force (SAC)
1	Strategic Air Command (OS)
3	Surgeon General
1	Sylvania Electric Products, Inc.
1	Technical Research Group
1	Tennessee Valley Authority
2	Union Carbide Nuclear Company (ORGDP)
7	Union Carbide Nuclear Company (ORNL)
1	Union Carbide Nuclear Company (Paducah Plant)
1	United Nuclear Corporation (NDA)
1	United Nuclear Corporation (OMC)
1	U. S. Geological Survey, Naval Gun Factory
1	U. S. Geological Survey, WR Division
1	U. S. Weather Bureau, Las Vegas
1	U. S. Weather Bureau, Washington
4	University of California, Berkeley
1	University of California, Davis
2	University of California, Livermore

EXTERNAL DISTRIBUTION (Contd.)

Number of Copies	
1	University of California, Los Angeles
1	University of California, San Francisco
1	University of Chicago, USAF Radiation Laboratory
1	University of Puerto Rico
1	University of Rochester
1	University of Tennessee (UTA)
1	University of Utah
1	University of Washington
1	Walter Reed Army Medical Center
1	Watertown Arsenal
1	Western Reserve University
2	Westinghouse Bettis Atomic Power Laboratory
1	Westinghouse Electric Corporation
1	Westinghouse Electric Corporation (NASA)
1	Yankee Atomic Electric Company
325	Division of Technical Information Extension
100	Office of Technical Services, Washington