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OUT-OF-PILE STUDIES OF FISSION-PRODUCT RELEASE FROM OVERHEATED REACTOR FUELS AT ORNL, 1955–1965

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OAK RIDGE NATIONAL LABORATORY
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for the
U.S. ATOMIC ENERGY COMMISSION

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JULY 1967

OAK RIDGE NATIONAL LABORATORY
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ABSTRACT

Studies of fission-product release from overheated reactor fuel materials started at ORNL in 1955 as a two-man effort to supply data needed to determine the hazard of nuclear reactor accidents. These studies, which have continued and expanded since that time, have generated a great deal of data on a variety of reactor fuels. Much of this information was reported in documents that received limited distribution. This report contains a discussion of factors affecting fission product release, a description of techniques developed in these studies, a compilation of data obtained in the 11-year period covered by the report, some illustrations of the use of the data, and some recommendations for further research. Other aspects of the out-of-pile fission product release studies, such as transport behavior of the released fission products, will be covered by future topical reports.

1.0 INTRODUCTION

Studies designed to provide information on the hazards of fission products resulting from destruction of reactor fuels in a nuclear reactor accident started as a part-time effort by a small group in the Chemistry Division of the Oak Ridge National Laboratory in 1955. Prototype power reactors were already under construction at that time and the only information available on the release of fission products from melted reactor fuels was a by-product of early efforts to develop pyrometallurgical methods for processing fuel materials.[1,2,3] Because of the scarcity of useful information on fission-product release from fuels, it was necessary, in order to evaluate the safety of early nuclear reactors, to assume[4] that 100% or a large percentage of the fission products would be released to the containment systems in nuclear reactor accidents. Results from early experiments conducted at ORNL[5,6,7,8] showed that such assumptions were overly pessimistic. This program has continued and expanded over the years, especially since 1961, to include in-pile release studies in the ORR[9] and TREAT[10] reactors, as reported in a recent symposium.[11] Interest has shifted from determinations of the extent of fission-product release from fuels to observations of the behavior of released fission products in simulated containment systems such as the Containment Mockup Facility,[12] and the Nuclear Safety Pilot Plant.[13] Interest extends also to determinations of the effectiveness of trapping systems[14] and to other proposed methods for diminishing the likelihood of escape of fission products from reactor containment systems, as well as to extensive investigations of methods of characterizing released fission products.[15] Even larger scale experiments will be performed at other installations.[16,17] Future topical reports from this laboratory will cover other aspects of nuclear safety research. Information on particles produced by overheated fuels will be included in the report on transport of fission products.

The results of the studies in some of the above-mentioned areas conducted at ORNL during the past 11 years under the guidance of one of the authors (G. W. Parker) have been published in a number of reports, some of which received limited circulation, and were described in speeches at a variety of meetings. (See Refs. 6, 12, 21, 50, 53, 54, 55, and 61.) This document was prepared to gather together the scattered information on out-of-pile fission-product release developed in this program, in order to make it more readily available. This report deals with studies of fission-product release resulting from the three primary release mechanisms, diffusion, melting, and oxidation. In order to interpret data on release accompanying the oxidation of irradiated fuels, it was also necessary to determine oxidation rates of these fuels and the results of these studies are also included in Sections 4-6 which are arranged according to fuel type.

For a more general coverage of the literature in this field, the reader is referred to recent reviews.[18,19] In this report, data published by other investigators are considered only for comparison with information reported here.

2.0 THEORETICAL CONSIDERATIONS

Nuclear reactor accidents are not susceptible to rigorous theoretical analysis but it is possible to recognize some of the factors that affect fission-product release. Mechanisms of release and the effect of free energy of formation of compounds and vapor pressure on the extent of release are discussed qualitatively.

2.1 Mechanisms of Release

2.1.1 Fuel Melting

The amount of a fission product released as a function of fuel melting temperature might be expected to follow a relatively simple relation between the fuel melting temperature and the vapor pressure of the fission product; however, since identical experimental conditions have rarely been maintained for different metals and alloys it has been difficult to establish the nature of the relationship. The release of some fission product elements would no doubt be affected by chemical reactions and by solubility in the fuel or its environs as well as by the fuel temperatures reached. Without fuel melting and subsequent release of fission products, a nuclear reactor accident would involve no more hazard to the general public than any conventional power plant accident of a similar nature. Thus melting, followed by the release of fission products, is one of the unique features of a serious nuclear reactor accident. The degree of melting is always difficult to postulate since much uncertainty exists concerning the rate of heat loss from the reactor core and the possibility of reassembly of a critical mass in the bottom of the primary vessel. The melting of fuels with reasonable burnup levels invariably leads to high release rates for the volatile elements (Xe, I, Te, Cs). The lowest melting fuels may also release ruthenium and cesium, in addition to xenon, iodine, and tellurium, while the higher melting ones may also release some strontium and barium.

2.1.2 Fuel Oxidation

Burning of either a metallic fuel (Section 4.1) or of a lower oxide (UO_2) (Section 6.1.2) greatly enhances fission-product release by increasing surface area by many orders of magnitude as well as by local overheating and gas expulsion. Hilliard[20] and Parker et al[21] have noted that the fraction of most fission products released from metallic uranium was proportional to the extent of oxidation. They also observed that the rate of release was nearly proportional to temperature up to 1500°C. One serious oversight which seems to exist in most hazards summaries is lack of consideration of the effect that UO_2 burning may contribute to the total release of fission products following meltdown or cladding rupture and subsequent exposure of fuel to air at temperatures below 1500°C. The correct procedure would be to sum the effect of melting or high-temperature diffusion with the effect of oxidation on the residual inventory of fission products.

2.1.3 Gaseous Fission Product Diffusion

Gas-phase diffusion by fission products at temperatures near or above their boiling point is the principal mode of escape from an unaltered fuel matrix. This process is enhanced by the effect of burnup. Bubbles or gas pockets become evident through swelling of the fuel upon heating just below the melting temperature. When the strength of the fuel is exceeded by internal gas pressure (dissolved gases or excess oxygen from UO_2), the bubbles break through the surface and sweep the collected gases including halogens and other volatile elements out of the fuel. At the tracer level, the release is often delayed and limited by slight solubility or by retention of fission products in lattice defects. At high burnup the release may begin below the fuel melting temperature as a result of cladding failure induced by the increase in pressure from accumulated rare gases.

The initial phase of the diffusion process (See Section 6.1.1) invariably consists of a prompt-burst-type release which may account for more than half of the total volatile release.[22] The residual fraction is released more slowly at a steady rate and the time for a given fraction to be released can be fitted to an equation of the type:

$$D = D_o e^{-Q/RT}$$

where Q is the energy of activation for the diffusion of the particular species that exists in the temperature range of interest. This latter process is of relatively little importance in nuclear safety considerations because, at the high temperatures required for rapid diffusion, another mechanism such as melting or oxidation is more likely to be the controlling factor. However, grain growth, a process occurring above 1700°C in UO_2, results in a large increase in diffusion rate (See Fig. 6.1).

2.1.4 Migration of Solid-Phase Fission Products

In heterogeneous fuel systems, solubility of the fission products in the fuel matrix and cladding is of little significance as a mechanism affecting release except perhaps at temperatures approaching the fuel melting point. Even partially melted fuel plates of aluminum alloy from a fuel melting accident in the Oak Ridge Research Reactor[23] showed no significant migration of fuel or cladding penetration in a part of the fuel plate very close to the melted region, presumably because such a process is strongly time dependent. Distinct similarities in physical properties of the fuel and fission product phases favor dissolution. For example, elements that alloy readily, such as tellurium, ruthenium, tin, and antimony, may dissolve in metallic fuel or cladding and thus result in an increase in migration through the cladding. Solubility in the sense of alloy formation may then favor retention by the fuel until the metallic fuel or cladding containing the alloyed fission products is completely destroyed by oxidation. Oxides of the rare earths or alkaline earths will dissolve in UO_2 at very high temperatures, thereby gaining additional access to the UO_2 surface and to the fuel-void volume. The alkaline earths, however, having relatively unstable oxides at high temperature, will volatilize rapidly if the system is extremely low in free oxygen as, for example, in the presence of melted Zircaloy cladding (see Table 6.9).

2.1.5 Compound Formation

Compound formation between fission products and fuel components is normally not significant; however, in theory it should occur to some extent between volatile elements such as cesium and the halogens when fuel rods are operated at temperatures high enough to permit distillation of these fission products and condensation in the cooler parts of the can. Such compounds, however, have relatively low stability and they would probably dissociate in the event of high-temperature cladding rupture. Experiments on fission-product release from uranium-aluminum alloy[24] failed to show appreciable compound formation when cesium and iodine were released simultaneously by melting, even though the aerosol of iodine and cesium was allowed to age for an extended period in the same container. This was demonstrated by the diffusion tube method.

Compound formation may be of somewhat more significance in the case of pyrocarbon-based fuels. The rapid diffusion of soluble but relatively unstable carbides of strontium and barium may account for the observed high rates of diffusion of these elements. The behavior of cesium, which also diffuses rapidly in graphite-matrix fuels, is affected by the formation of interlamellar compounds, such as CsC_8.[25] Other fission products (e.g., zirconium-niobium) are immobilized as carbides because of the high temperature stability of the compounds. It seems likely, however, that compounds which form when two fission-product elements are deposited on the same surface may be of more importance than those formed in the fuel.

It has been reported[26] that there is evidence of formation of a uranium iodide compound when irradiated uranium is melted in pure helium. Since fission-produced iodine atoms are surrounded by uranium atoms, it appears that favorable conditions for reaction exist. Uranium iodides are easily oxidized and it is highly unlikely that fuel materials will be surrounded by pure, non-oxidizing gases in a reactor accident, hence it seems probable that uranium iodide formation would not significantly affect release of fission-product iodine from uranium or uranium alloys under accident conditions.

2.2 Relation of Free Energy and Vapor Pressure to Fission Product Release

2.2.1 Oxide Fuel Systems and the Effect of Oxygen on Fission-Product Release

Processes favoring release of fission products to the environment and those favoring retention in the fuel were discussed by Parker et al.[8] Data in this same report indicated a correlation between release of rare gases or iodine and fuel melting temperature. The results of studies performed subsequent to that report, especially on the release of fission products from high burnup fuel materials, have altered some of the earlier beliefs on the subject but there can be little question that the chemical form of fission products has a profound effect on their release from fuel during reactor accidents and on their subsequent behavior. Methods of determining directly the chemical form of released fission products remain to be devised but it is possible to draw inferences as to their probable chemical form based on observations of the effect of varying environments on the extent of fission-product release.

The two physical properties most relevant to this subject are the vapor pressure of the elements and compounds that can form under accident conditions and the free energies of formation of the latter, which indicate the stability of the compounds at elevated temperatures. It should be recognized that thermodynamic data apply, strictly speaking, only to equilibrium conditions which seldom, if ever, exist in reactor accidents. Consequently, it is necessary to use caution in applying such data to prediction of the behavior of fission products under accident conditions, but it seems probable that, at elevated temperatures resulting from loss-of-coolant accidents in reactors fueled with high-melting materials, equilibrium will be at least approached so that conclusions based on thermodynamic considerations are of some value. Kingery and Wygant[27] have discussed other limitations on thermodynamics with special reference to ceramic materials. The low concentrations of fission products in fuels irradiated to burnup levels expected to be attained in most power reactors makes the assumption of ideal behavior seem reasonable. Discussion of fugacities seems unwarranted because they provide second order refinements which are not significant considering the large uncertainties and uncontrolled variables existing in reactor accidents.

The free energy of formation of oxides is of considerable importance in predicting the form of released fission products because UO_2 and $(U,Th)O_2$ fuels are being used or proposed for use in a large number of nuclear-power reactors and, also, because oxygen is likely to be present in the environment of accident-ruptured fuel materials. Glassner[28] has assembled a useful compilation of thermodynamic data on oxides as has Coughlin.[29] Similar data for the elements have been tabulated by Stull and Sinke[30] and by Kelley and King.[31] Unfortunately, it is usually necessary to extrapolate available data considerably to obtain free energy values at the melting point of UO_2 ($3100^{o}K$) or at higher temperatures. Extrapolations or estimates are usually sufficiently accurate to indicate whether or not a fission product oxide is stable at temperatures of interest. For example, the free energy of formation of Cs_2O is positive at temperatures above approximately $1600^{o}K$ showing that elemental cesium is likely to exist in the vapor phase at high temperatures even in air and thus its release is likely to be relatively unaffected by the presence of oxygen. It will react readily, of course, with oxygen at lower temperatures so that its behavior subsequent to its release may be affected to a greater extent by the environment than is its release.

Free energy data also indicate that RuO_4 is stable at temperatures up to $1700^{o}K$. Hilliard and Reid[32] postulated the formation of this highly-volatile oxide (B.P. 135°C) to account for ruthenium-release values found when high-burnup uranium specimens were oxidized in air. These investigators say that RuO_4 is apparently not formed in the presence of uranium metal or lower uranium oxides. The behavior of molybdenum released from oxidizing uranium appears[32] to be similar to that of ruthenium, although free energy values show that its oxides are more stable than the ruthenium oxides. The reported[32] decreasing release of tellurium with increasing temperature, ascribed to the decreasing stability of TeO_2, was not confirmed by the results of similar studies.[21] Recently reported vapor pressure data[33] show that tellurium is more volatile than TeO_2.

A compilation of vapor pressure data similar to that given earlier[8] is shown in Fig. 2.1 and Fig. 2.2. Based on the information shown in these figures, one could predict, for example, that the release of barium and strontium will be much greater in the absence of oxygen than in its presence, since the elements are more volatile than the oxides and the free energy values[28] also show that the oxides are quite stable at high temperatures. There is abundant support for this prediction in the fission-product-release literature.

2.2.2 Graphite-Based Fuels

Interest in the use of fuels containing uranium carbide in a graphite matrix in high-temperature gas-cooled reactors has prompted both theoretical and experimental studies of fission-product behavior in fuel materials and coolant streams. Two theoretical studies were reported by Brewer.[34,35] These brief reports, which were prepared primarily for internal use at General Atomics, unfortunately do not specify the fuel composition or the burnup level considered and they also give no information on the methods employed in the calculation of results reported but they contain some interesting conclusions

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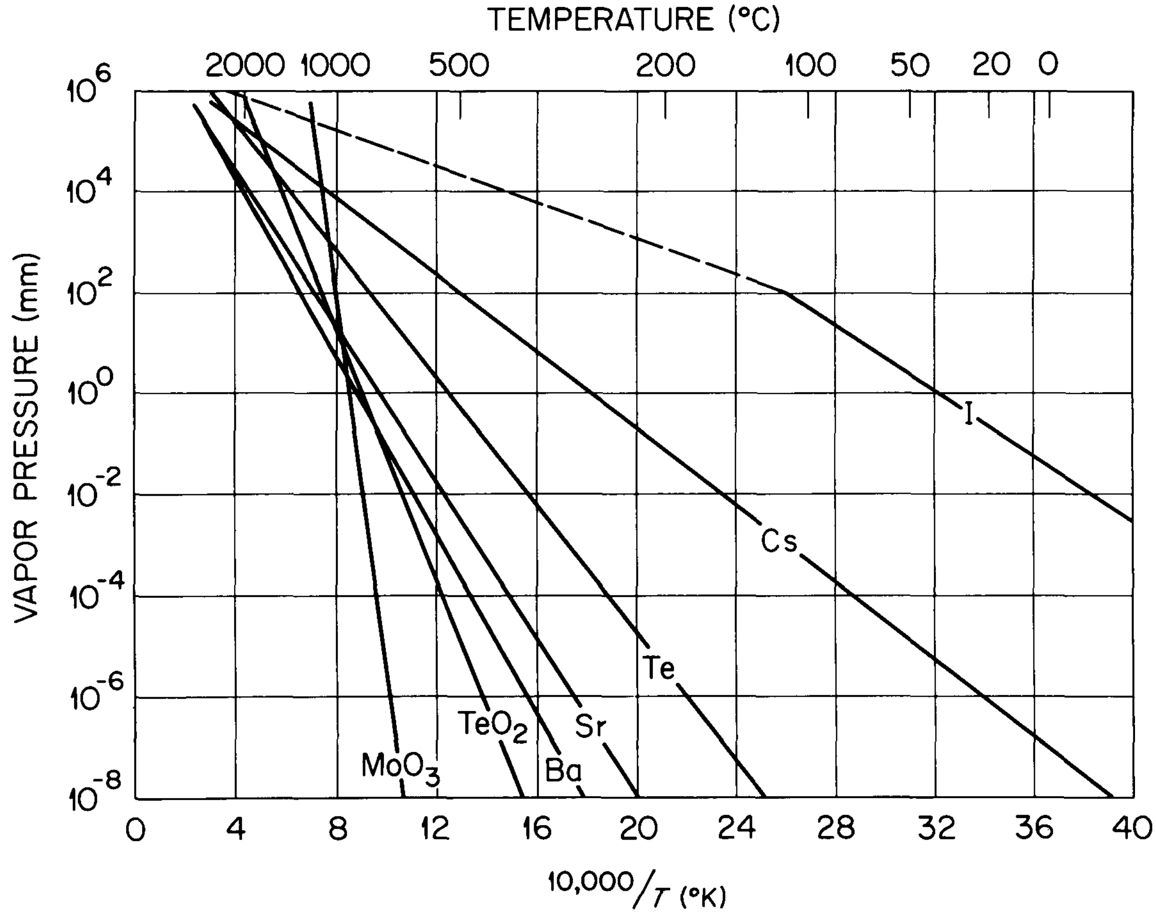

Fig. 2.1. Vapor Pressures of Volatile Elements and Oxides.

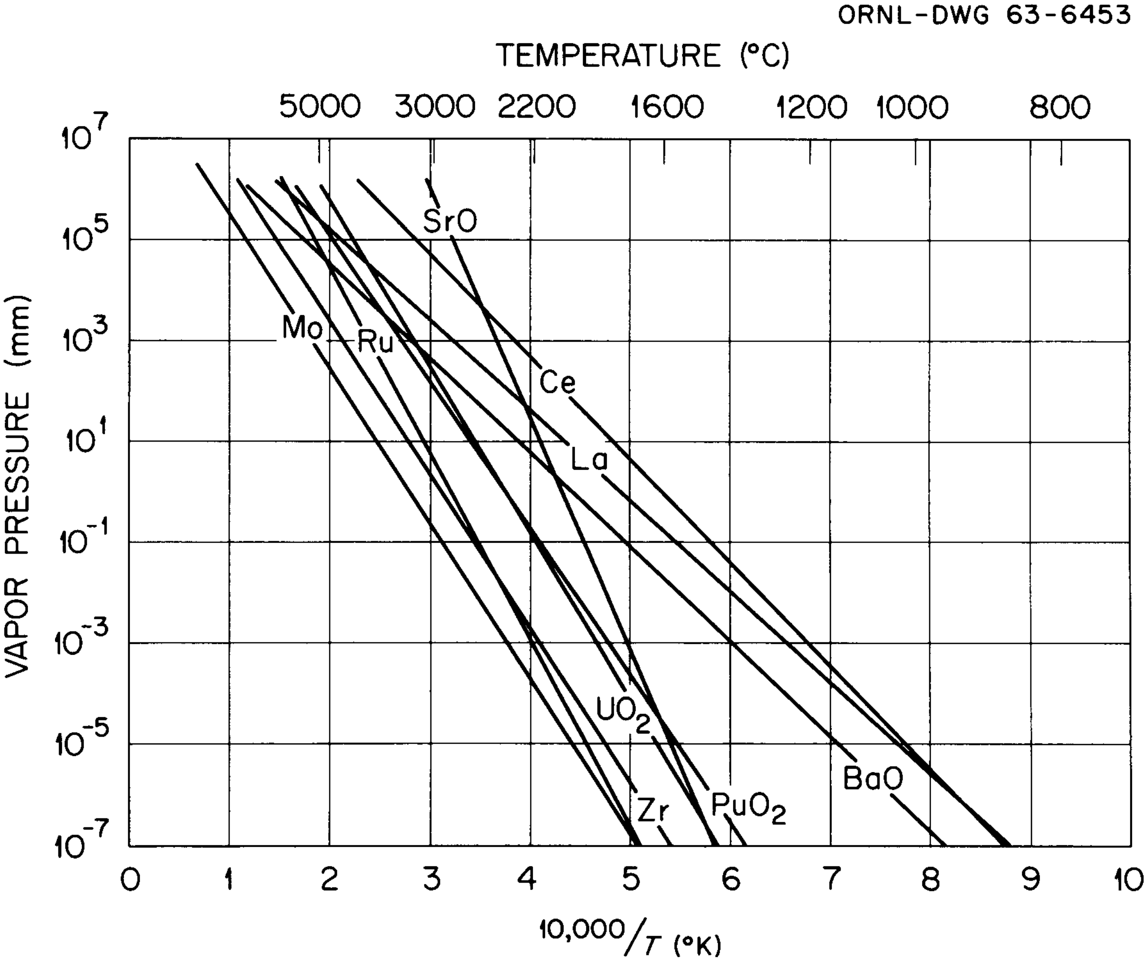

Fig. 2.2. Vapor Pressures of Low-Volatility Elements and Oxides.

based on thermodynamic and vapor pressure data. Thermodynamic data on carbides compiled by Brewer, et al.[36] have been revised and extended by Krikorian.[37,38] Very little data on the vapor pressure of carbides exists but, since most of the fission-product carbides are unstable at high temperatures, the vapor pressures can be assumed to be those of the elements.

Brewer[34,35] postulates that fission-product bromine and iodine will combine with fission-product cesium (or rubidium). Experimental evidence supporting this belief is lacking at the present time. Brewer predicted[35] that, in a "runaway" reactor containing fuel elements brazed with ZrC, the upper temperature limit of fuel-can integrity would probably be set by the melting temperature of the ZrC-C eutectic which has been reported[39] to be 2430°C. He also indicated that internal gas pressure (36 atm due to fission products alone, assuming no condensation) would probably cause can failure before it reached this temperature. He states that the vapor pressure inside the fuel can, after "extensive" burnup, will be between 40 and 60 atmospheres, including the helium pressure, at the melting temperature (2500°C) of the carbides.

3.0 EXPERIMENTAL TECHNIQUES

The equipment and techniques used in out-of-pile studies of the release of fission products from fuel materials will be discussed in the approximate order of historical development.

3.1 Low-Frequency Induction Heating Apparatus

An induction heating arrangement using graphite cylinders as the susceptor material was used in early fuel melting experiments[8] to obtain high temperatures, as shown in Fig. 3.1. The fission-product collection train differed from that

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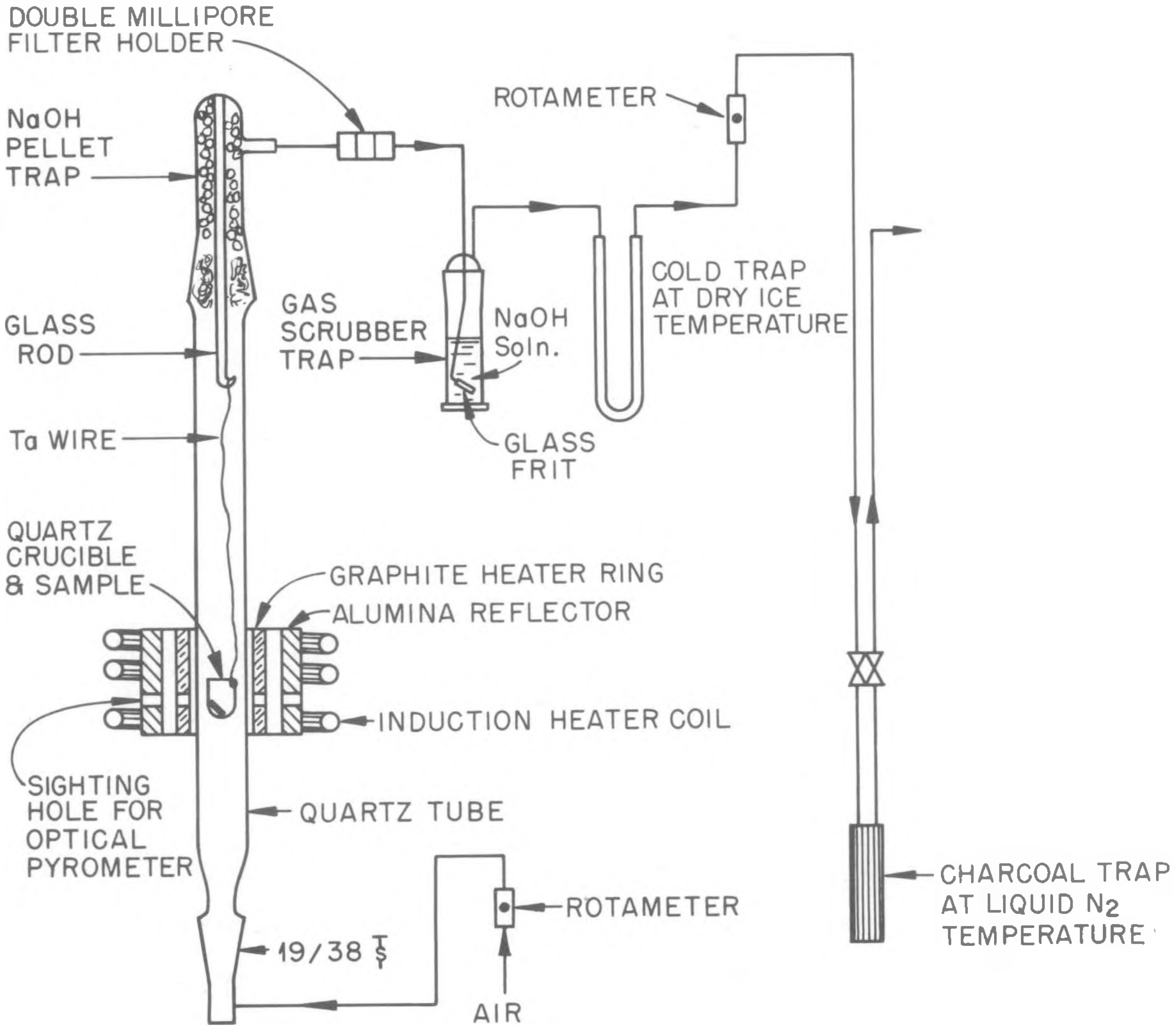

Fig. 3.1. Induction Heating Furnace and Fission-Product Traps for Air Melting Experiments.

discussed in the following sections. Sodium hydroxide pellets and a sodium hydroxide scrubber solution were used primarily to collect released iodine. Part of the airborne cesium was collected by the pellets and the remainder was trapped in the Millipore filters. Rare gases were collected in the cold charcoal trap.

Temperatures were measured by use of an optical pyrometer and variations in this parameter were effected by changing the thickness of the graphite susceptor. The power source used with this apparatus was a Federal Telephone and Radio Corporation Megatherm unit.

3.2 Apparatus for Induction Heating of Metallic Fuel in Steam

Induction heating of fuel specimens in steam was accomplished with the same equipment described in Section 3.1. A schematic diagram of this apparatus[8] is shown in Fig. 3.2. The steam flow rate was controlled by the gas flow rate and the temperature of the boiler heater. In this arrangement, the metallic fuel served as the susceptor. The potassium permangate trap was inserted ahead of the hot charcoal trap for the purpose of oxidizing any gaseous iodides to produce molecular iodine.

3.3 Apparatus for Determining Fission-Product Release During Oxidation and Melting of Uranium and Other Metallic Fuels

Equipment used in early oxidation and release studies[21] is shown schematically in Fig. 3.3. The platinum resistance furnace was preheated to any desired temperature up to 1550°C in the position shown with helium flowing through the Mullite furnace tube and, to start the oxidation, air or other oxidizing gas was introduced shortly after the furnace was moved

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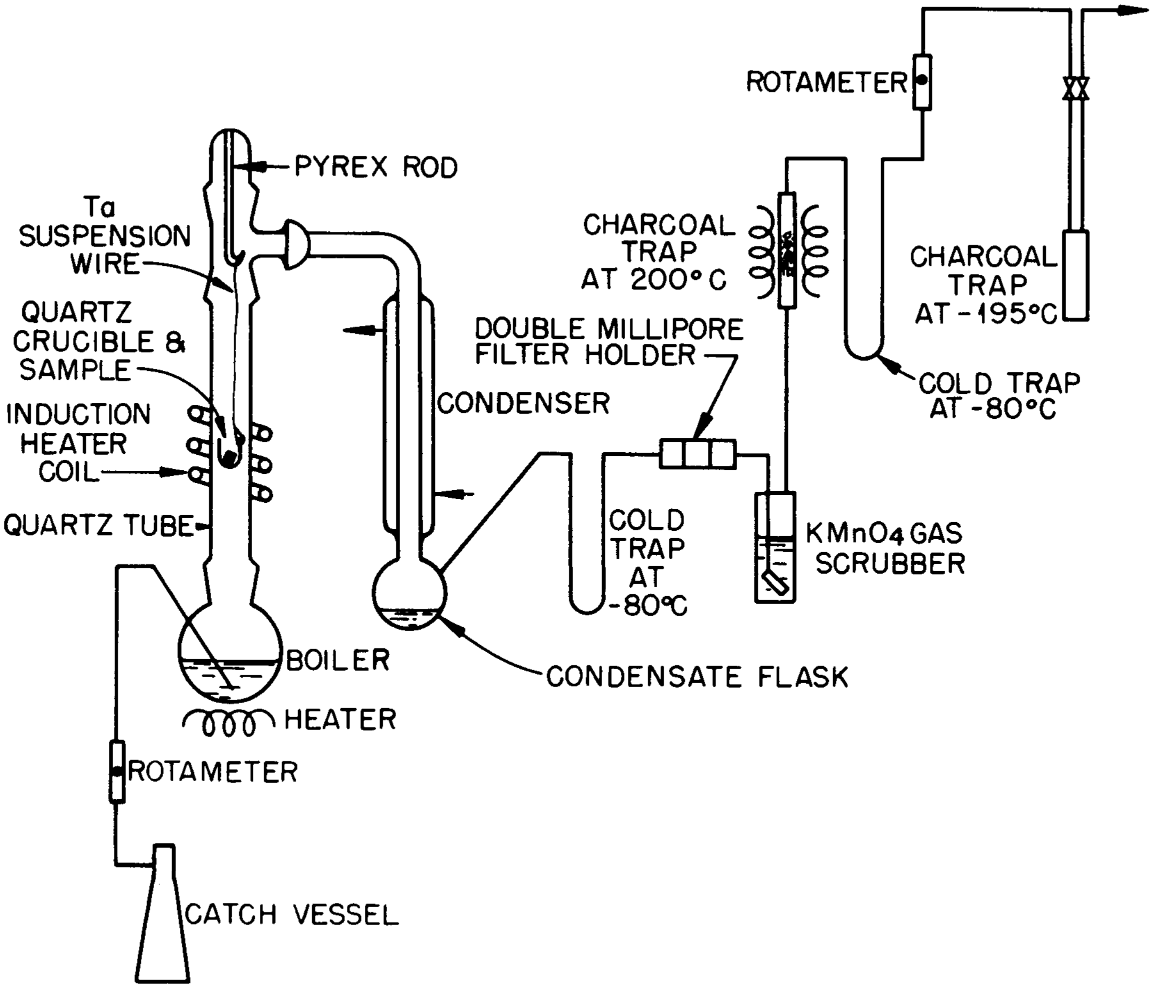
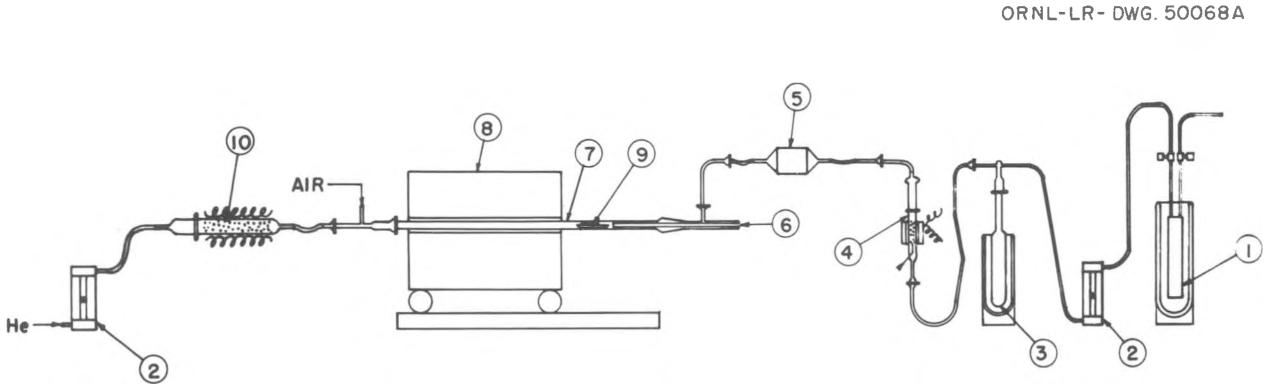


Fig. 3.2. Induction Heating Furnace and Fission-Product Traps for Melting Fuel Capsules in Steam.



1 — CHARCOAL TRAP AT -195° C
2 — ROTAMETER
3 — COLD TRAP
4 — CHARCOAL TRAP AT 250° C
5 — MILLIPORE FILTER HOLDER
6 — THERMOWELL
7 — QUARTZ COMBUSTION TUBE
8 — Pt RESISTANCE FURNACE
9 — COMBUSTION BOAT AND SAMPLE
10 — HOT Cu TURNINGS

Fig. 3.3. Schematic Diagram of Fission Product Release Apparatus.

around the sample. After a predetermined period, the reaction was stopped by substituting helium for the oxidizing gas and rolling the furnace back to its initial position to permit rapid cooling of the fuel material. To determine the rate of oxidation, the unoxidized uranium was dissolved in HCl and the U^{+4} was titrated with a standard dichromate solution to determine the fraction of uranium oxidized. Airborne particles were collected by Millipore filters, iodine in the heated charcoal trap, and rare gases in the liquid-nitrogen cooled charcoal trap.

Fission-product release was determined as follows. The apparatus was disassembled after cooling to room temperature. The furnace tube and the tubing connections to the Millipore filter holder, as well as the holder itself, were leached with a sodium hydroxide solution, dilute nitric acid, and an ammonium fluoride solution. The fuel residue was dissolved along with any released fission products that may have remained in the boat and the Millipore filters were likewise dissolved. The volume of all solutions was measured and aliquots were submitted for radiochemical analysis. The amount of iodine collected by the hot charcoal beds was determined by a counting technique and the amount of rare gases collected by the cold charcoal bed was compared to the amount collected during dissolution of the fuel residue by inserting the traps in an ionization chamber. Radiochemical analysis of the hot charcoal material was performed when there was reason to believe that fission products other than iodine or rare gases had penetrated the Millipore filters. This occurred only rarely when the filters were intact.

3.4 Thermobalance Apparatus

Rate of oxidation data could be obtained more conveniently than with the equipment shown in Fig. 3.3 by use of the thermobalance apparatus[21] shown in Fig. 3.4, arranged for oxidation

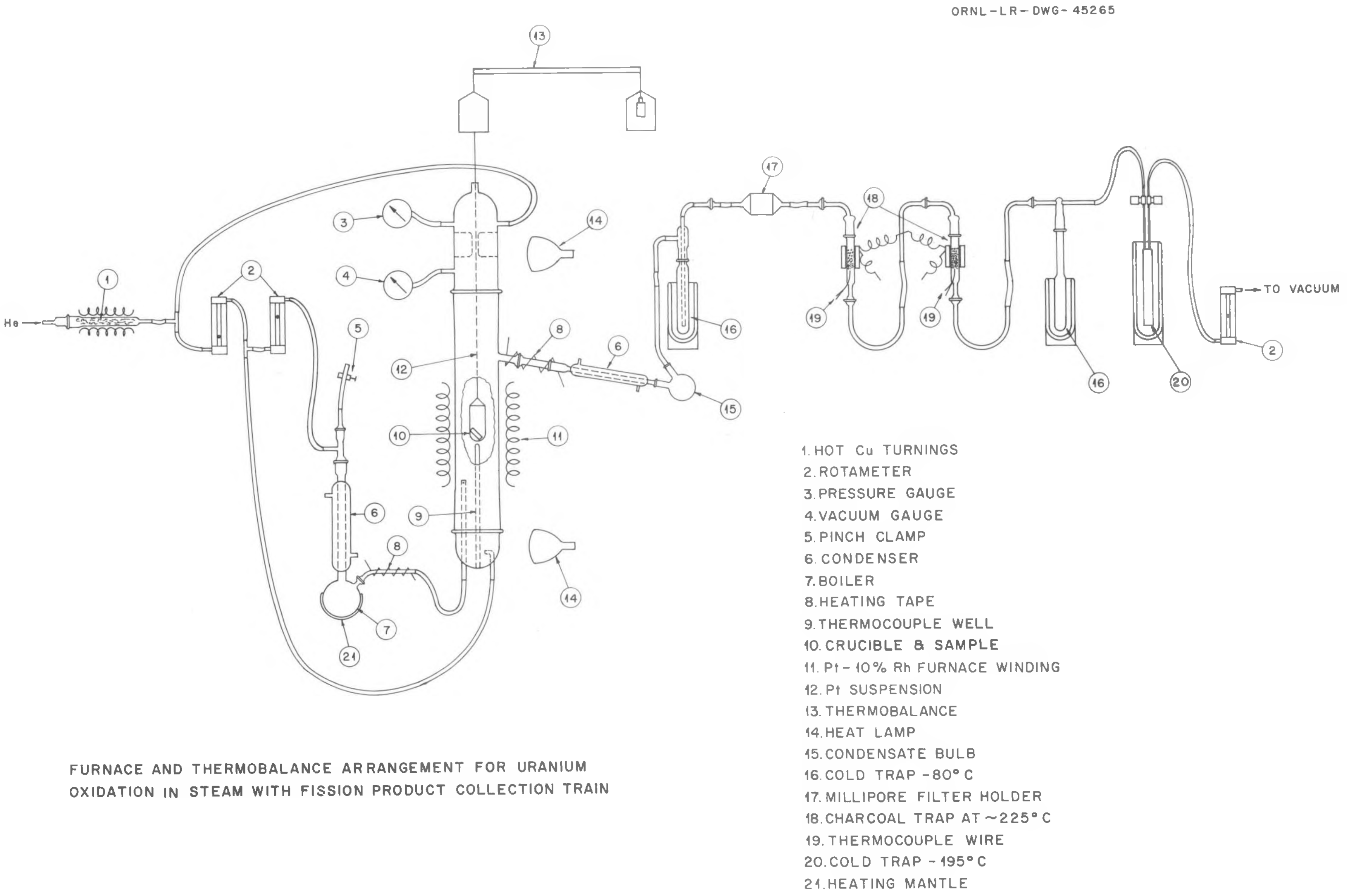


Fig. 3.4. Furnace and Thermobalance Arrangement for Uranium Oxidation in Steam with Fission Product Collection Train.

with steam. A Mauer Recording Thermobalance (Niagara Electron Labs, Andover, N. Y.) was used to weigh fuel specimens during exposure to oxidizing atmospheres. The suspension wire from the balance passed through a constriction between two pressure gauges having a range of 0 to 1-in. of water. Helium was introduced through the top opening at a rate of about 150 cm^3/min to give a positive pressure of 0.4-in. while a vacuum applied to the exit end of the fission-product collection train, following the cold charcoal trap, gave a negative pressure of approximately 0.1-in. at the lower opening. Thus a controlled leak of helium into the furnace was maintained to insure containment of fission-product gases and exclusion of air during oxidation or melting experiments. The principal problems in operating this equipment were in obtaining the proper alignment of the furnace tube to avoid contact with the sample suspension wire and the necessity of close proximity of the experimenter to the radioactive fuel while it was being transferred into or out of the apparatus. The balance, furnace, and fission-product collection train were placed in a shielded hood that minimized exposure to radioactivity while experiments were in progress. Experiments involving use of steam presented the additional difficulty of avoiding condensation before the gas stream reached the exit condenser. The fission-product collection train was essentially the same as in Fig. 3.3, except for the condenser and additional cold trap required by the steam atmosphere. The procedure for determining the amount of released fission products was the same as that described in the previous section, except that the condensate was also analyzed.

3.5 Arc-Image Furnace for Melting UO_2 and Other Ceramic Fuels

The arc-image furnace (Arthur D. Little Co.) shown in Fig. 3.5 was used in early fuel melting experiments[40] with

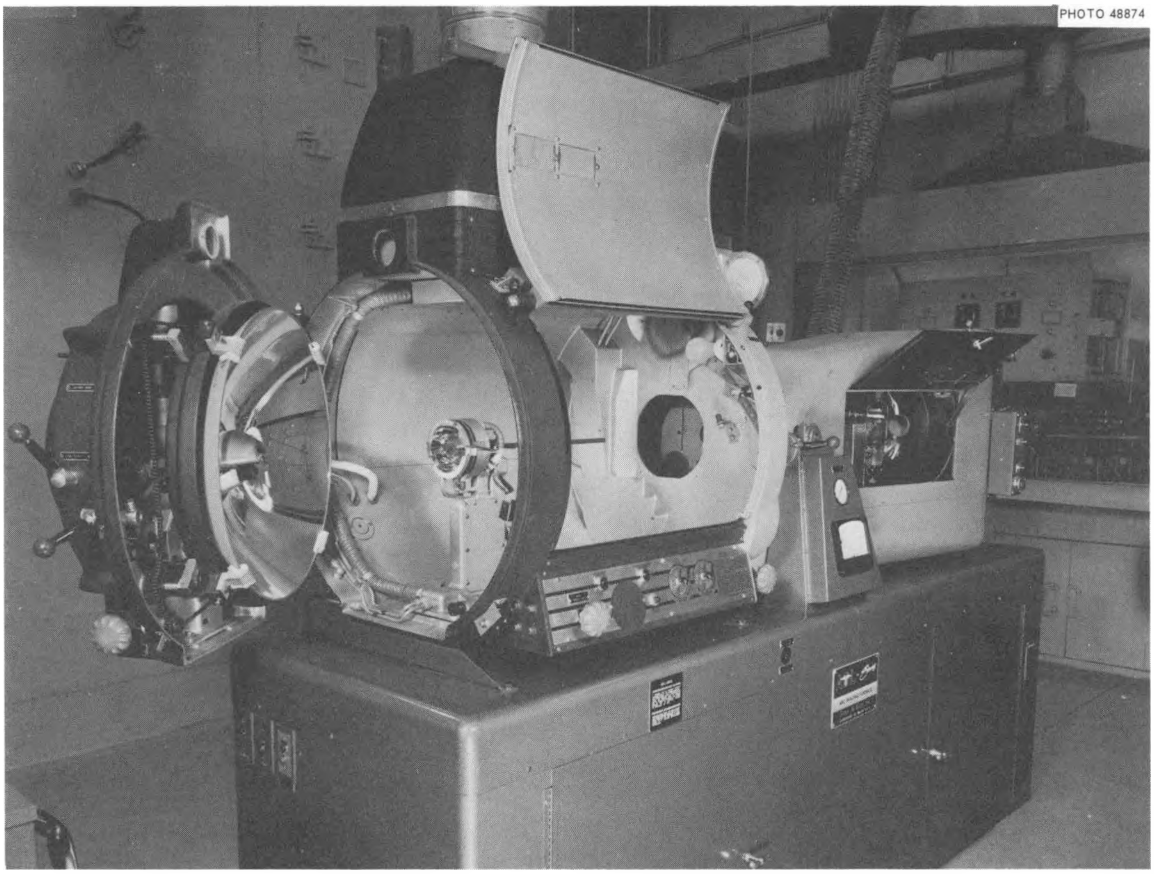


Fig. 3.5. Arc-Image Furnace.

uranium dioxide and BeO-UO_2 fuel materials. The photograph was made before the furnace was enclosed by a lead shield for work with high-burnup fuel material. A schematic diagram of the furnace and the fission-product collection train is shown in Fig. 3.6. Very high temperatures could be achieved in a small (1-cm diameter), very shallow volume. Accurate focussing of the arc was facilitated by a small light source in the arc position. The position of the fuel holder could be changed while the arc was on, in order to keep the unmelted portion of the specimen in focus. This apparatus worked quite well for melting small, cylindrical or octagonal shaped BeO-UO_2 specimens irradiated to tracer levels or for melting 25 to 60 mg samples of high burnup UO_2 enclosed in a BeO cylinder which was melted along with the fuel.

3.6 Induction Furnace and Apparatus for Release of Fission Products by Diffusion From UO_2 in a Helium Atmosphere

Apparatus for heating 30-g amounts of tracer-level-irradiated UO_2 in flowing helium is illustrated by the schematic diagram, Fig. 3.7. The tantalum crucible containing UO_2 powder served as the susceptor in this arrangement and this was surrounded by an alumina reflector.[41] Temperatures were measured by sighting down into the crucible with an optical pyrometer. Helium was purified by passage over hot zirconium rather than hot copper as in earlier apparatus. The exit gas passed through a trap filled with NaOH pellets, a hot charcoal trap, and then through cylindrical chambers partially occupied by Geiger tubes that gave a continuous indication of the amount of radioactive gas released from fuel. Finally, the krypton and xenon gas was collected by a liquid-nitrogen-cooled charcoal trap. The appearance of the tungsten crucible and melted UO_2 specimens is shown in Fig. 3.8. The thermocouple well at the bottom of the crucible

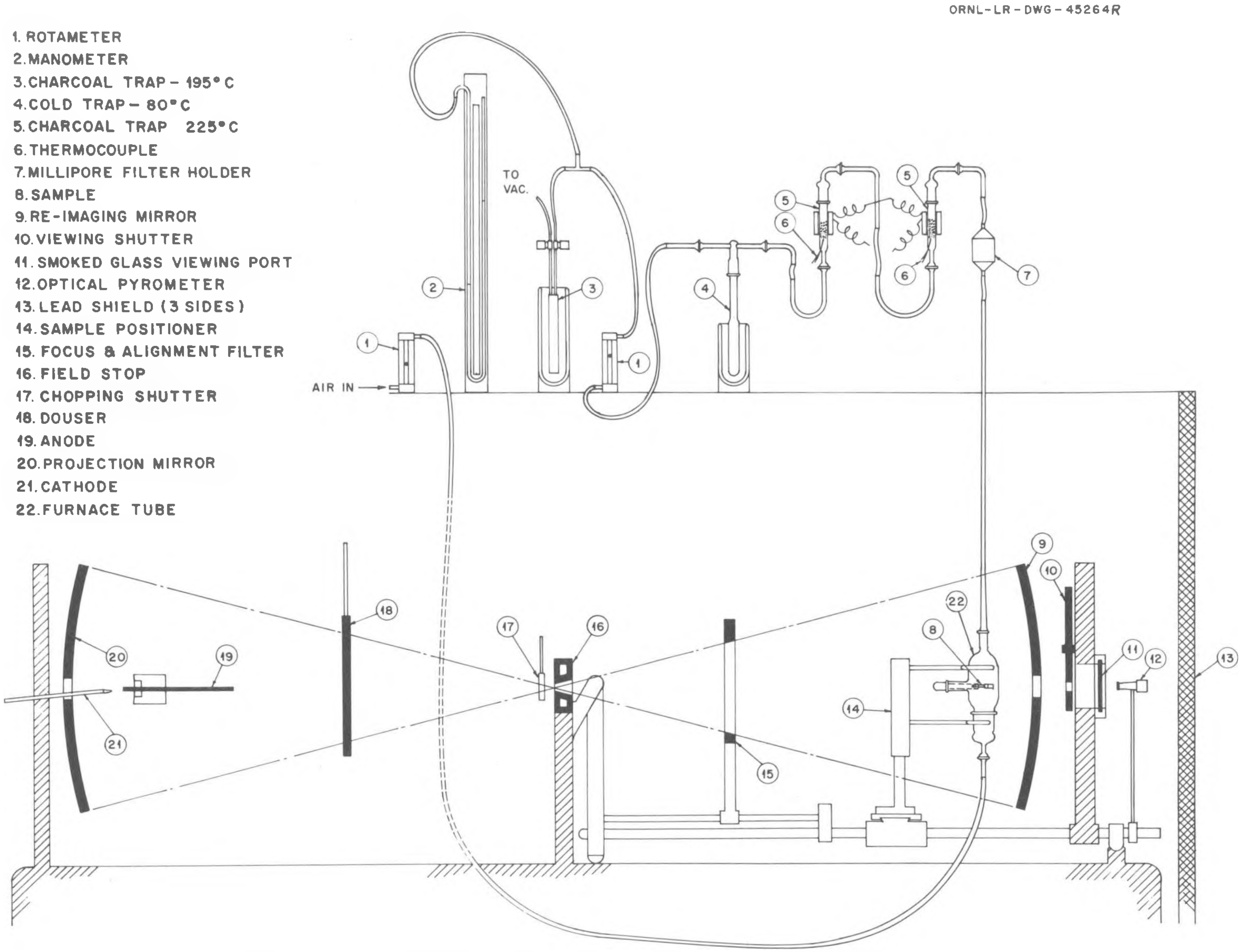


Fig. 3.6. Schematic Diagram of Arc-Image Furnace and Fission-Product Collection Train.

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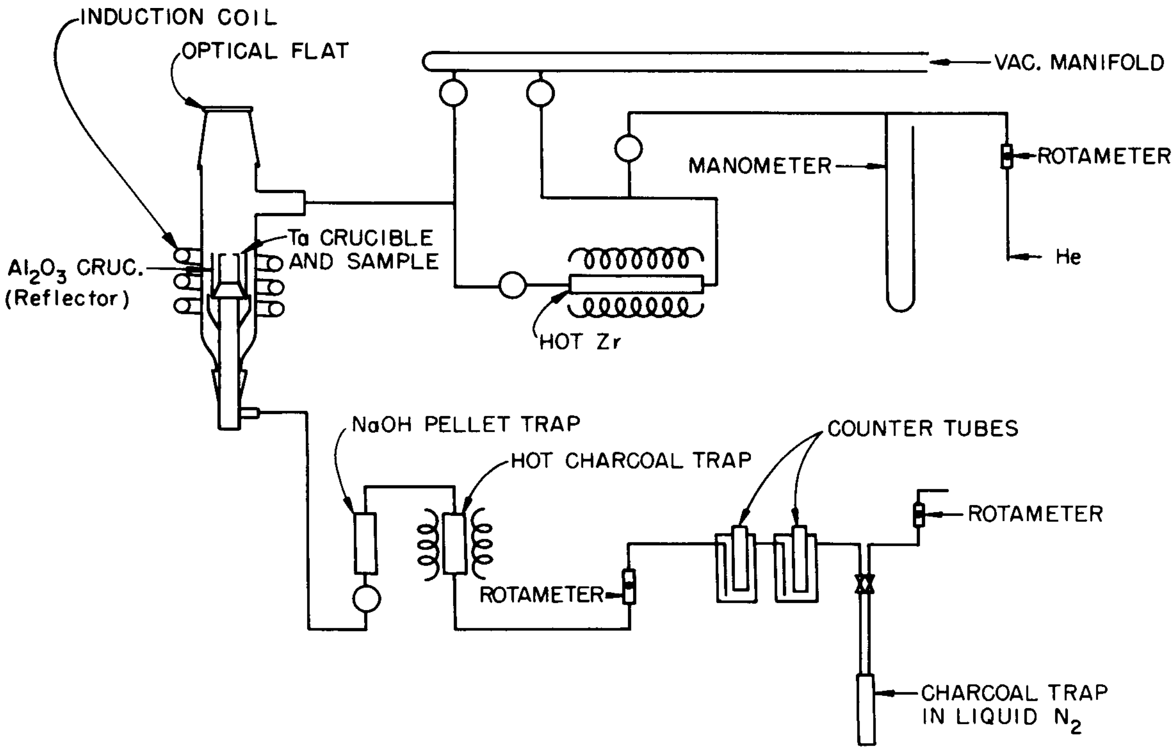

Fig. 3.7. Induction Furnace and Apparatus for Fission-Product Diffusion Studies with UO_2 in Helium.

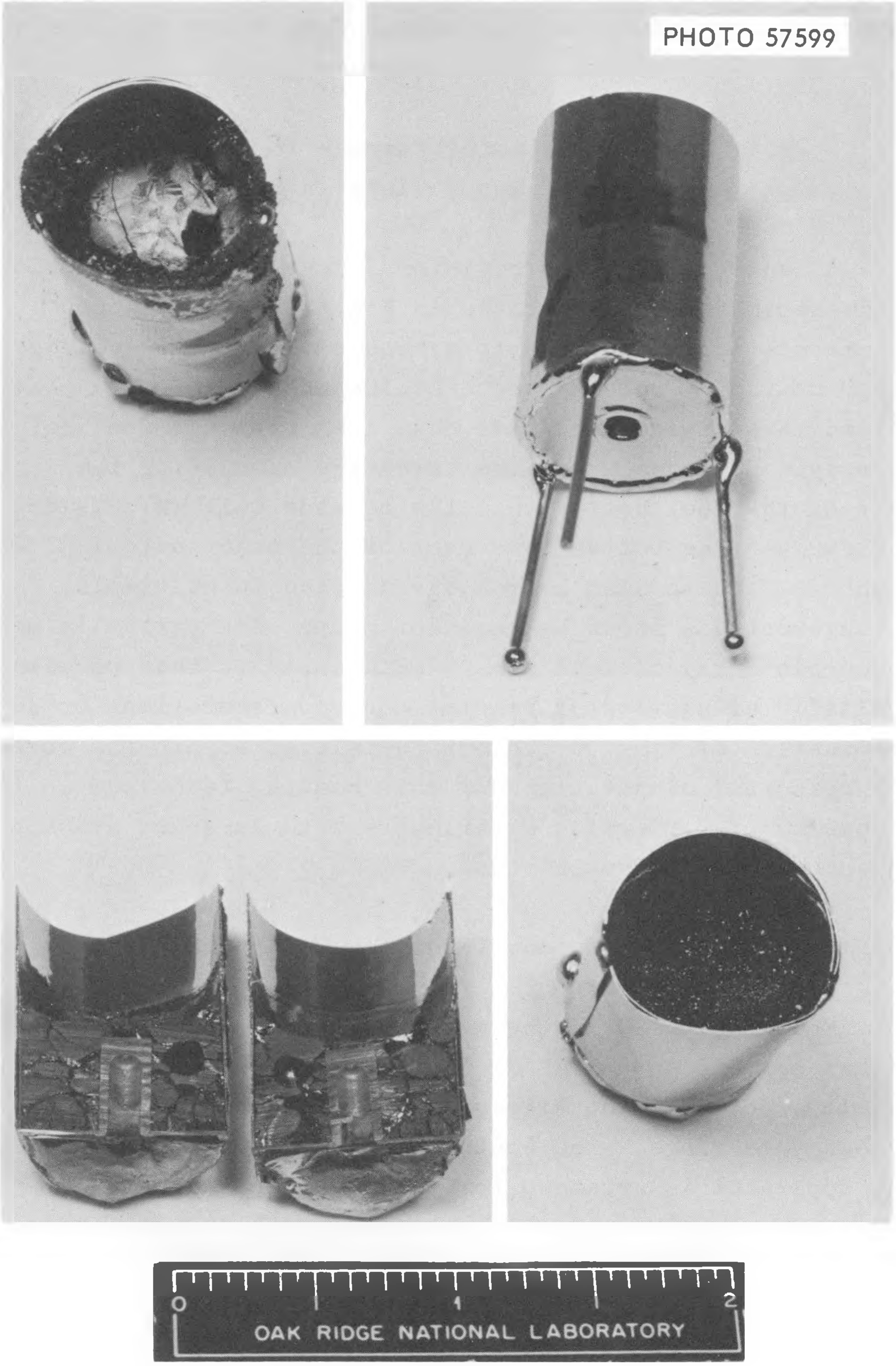

Fig. 3.8. Tungsten Crucibles with Melted UO_2 Specimens.

accommodated a tungsten-tungsten 26% rhodium thermocouple in some experiments. Oxidizing atmospheres cannot be used with this apparatus because of the tungsten crucible.

3.7 Tungsten Resistor Furnace for Melting Uranium Dioxide Pellets in Helium

A centered tungsten resistor[41] passing through hollow UO_2 fuel specimens is illustrated in Fig. 3.9. The copper electrodes were connected to a high-amperage, low-voltage power source. Tungsten heat shields and the alumina container helped to minimize radiative heat loss from the fuel material. This type of furnace has the advantage of heating the inner part of the fuel hotter than the outside wall which simulates nuclear heating better than most of the other out-of-pile methods. It can also be readily adapted to multi-pin arrangements as shown by the photograph of a partially melted seven-pin array of fuel pins[42] (Fig. 3.10). This permits realistic simulation of reactor core configuration for determination of fission product deposition within the core. The principal disadvantage of this heating technique is the necessity of performing experiments with an inert atmosphere because of the tungsten rods.

3.8 Dual Frequency Induction Heating Apparatus

Equipment used in the most recently developed heating technique[43] is shown in Fig. 3.11. The induction heater develops 50 kw in the kilocycle range and 25 kw in the 2 to 5 megacycle range. The control unit is in the foreground. The apparatus is arranged for tests with power leads of the length needed for hot cell operations with the oscillator located outside the cell. These tests indicated that it was difficult to transmit the required power with leads of the length shown and the oscillator unit, just behind the control

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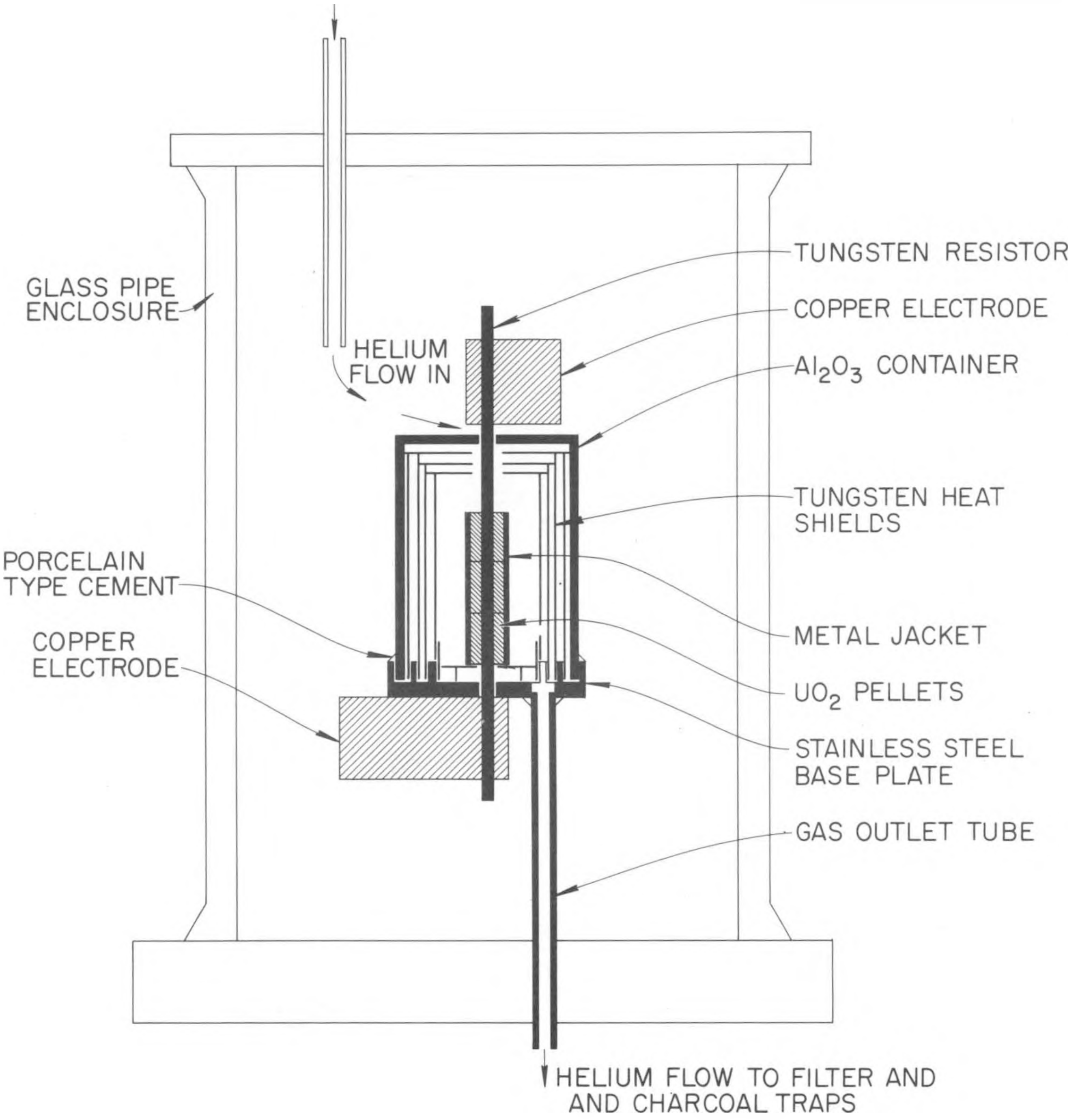

Fig. 3.9. Single Element Tungsten Resistor Furnace for Melting UO_2 Pellets in Helium.

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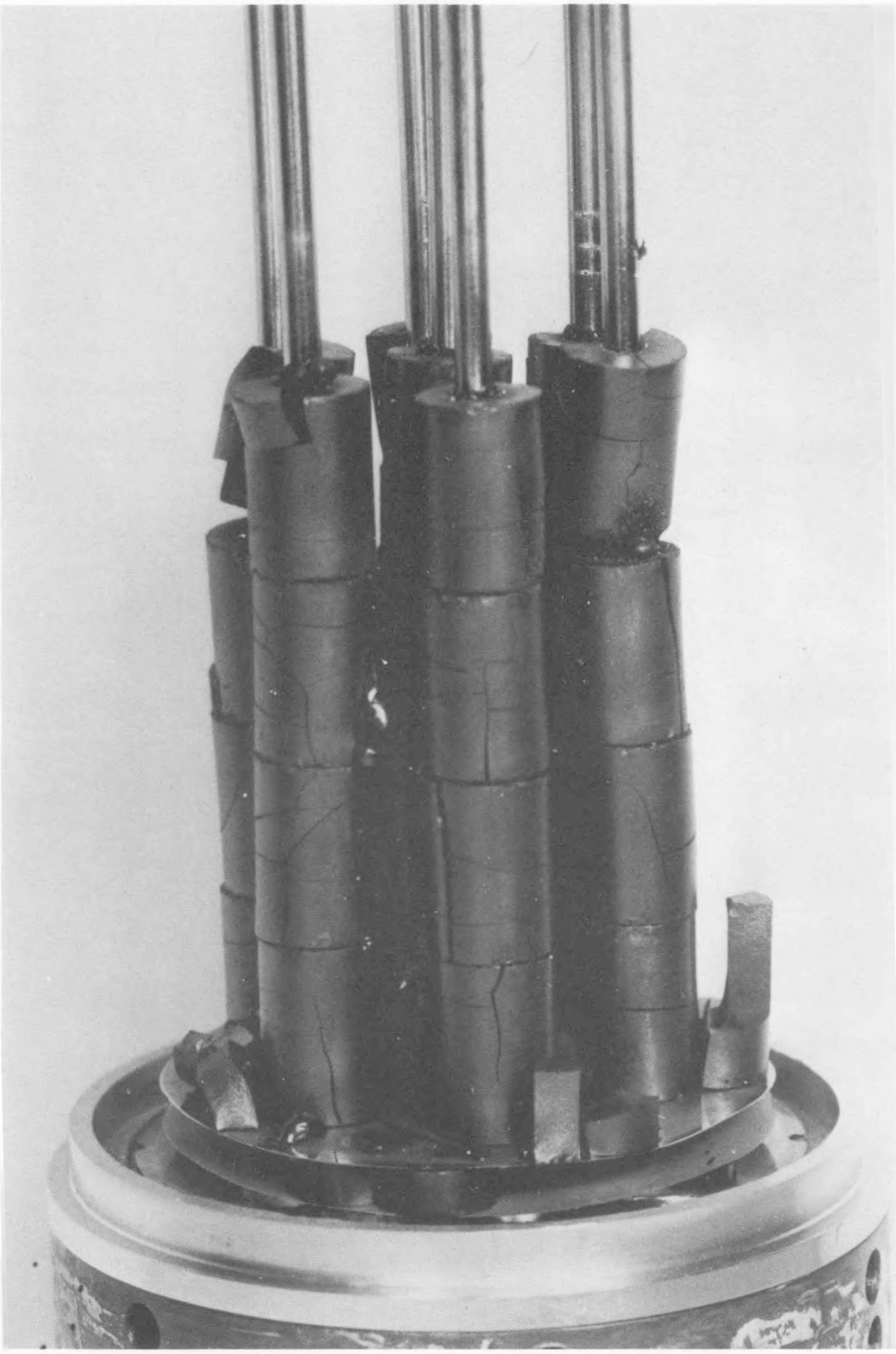

Fig. 3.10. Partially Melted UO_2 Fuel Heated by the Center-Resistor Method.

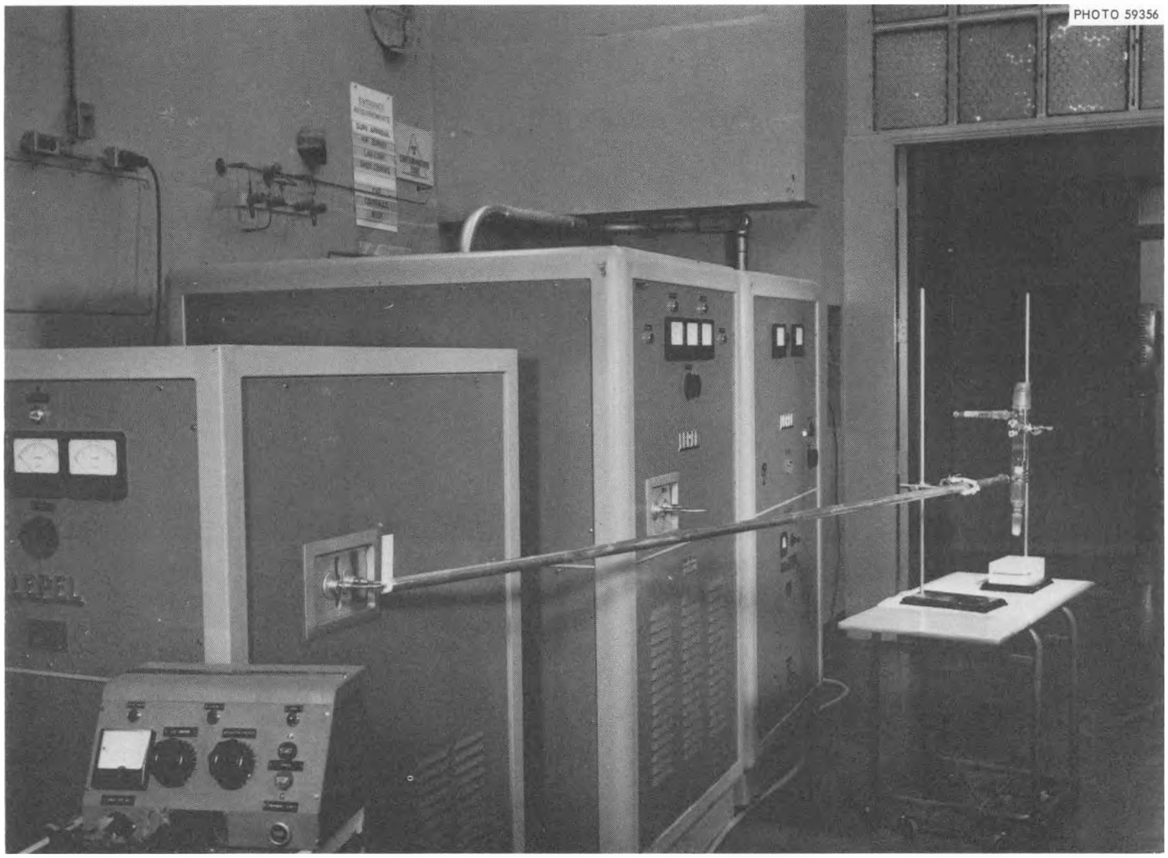

Fig. 3.11. High-Frequency (5-Mc) Induction-Heating Power Source for Melting Clad UO_2 by Direct Coupling.

unit in Fig. 3.11, was later placed inside the hot cell.

The motivation for purchase of the dual-frequency induction heater was to provide the capability of coupling with the metal cladding of UO_2 fuel specimens with the low frequency unit and then to couple to the UO_2 itself with the high frequency heater after the cladding melts because UO_2 becomes conducting at high temperatures. In practice, it has been found practical to couple with the cladding at high frequencies so that the kilocycle unit is not actually required.

3.9 Pressurized Induction Heating Furnace

The induction heating equipment discussed in the previous section has been used for melting clad UO_2 specimens in the pressurized furnace[44] shown in Fig. 3.12. It is necessary to have a pressurized furnace because released fission products are carried into a tank filled with a mixture of steam and air, usually at about 30 psig. The inner quartz tube supports a quartz boat partially filled with granular UO_2 on which the clad UO_2 specimen rests. The outside of the tube is air cooled and the chamber is at a higher pressure so that if the inner tube is ruptured, gas flow will be inward to prevent escape of fission products. The furnace is designed for remote loading of highly radioactive fuel materials.

4.0 INVESTIGATIONS WITH METALS AND ALLOYS

4.1 Oxidation of Metallic Uranium

The importance of experimental studies of the extent of fission-product release accompanying the oxidation of irradiated uranium in different atmospheres was demonstrated by the historic Windscale incident.[45,46] Reactors fueled

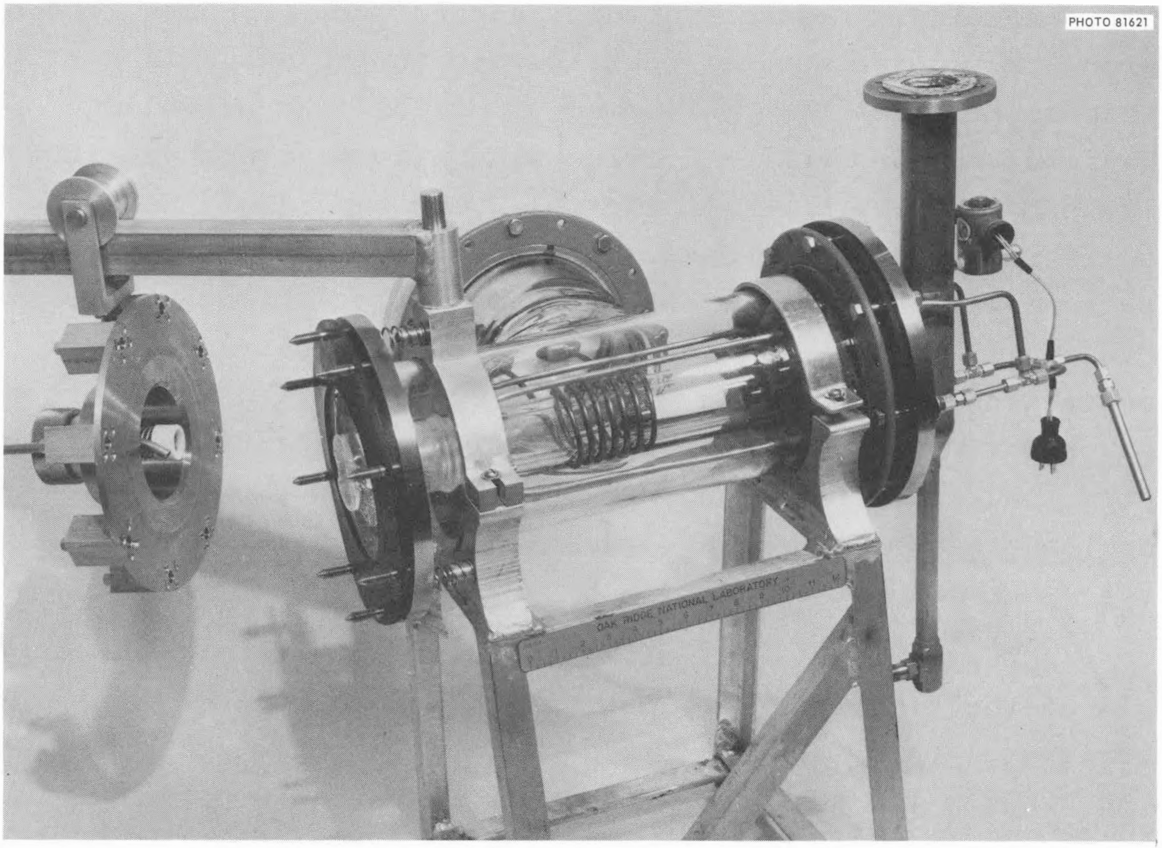

Fig. 3.12. Pressurized Induction Heating Furnace.

with metallic uranium occupy a minor power-production role in the United States, but they continue to be of importance in plutonium-producing reactors in this country and abroad, as well as in gas-cooled power reactors, principally in Great Britain. Loss of coolant in reactors of this type may result in hot metallic uranium coming in contact with air, steam, or CO_2, creating a reactor accident hazard from released fission products. Other studies contributing to the evaluation of this hazard have been performed principally at Harwell, Hanford, and Brookhaven. Some publications from these installations are referenced in the following sections. Since it seems reasonable to assume that rates of fission-product release will be proportional to rates of oxidation, and experimental studies have established the validity of this assumption, a discussion of the effect of various parameters on oxidation rates is important for an understanding of this potential reactor hazard. The equipment and procedures used in the present studies were described in Sections 3.3 and 3.4.

Experiments with uranium irradiated at trace level and all of the "incomplete-oxidation" runs were conducted in the horizontal tube furnace (Fig. 3.3). In experiments where the uranium was not completely oxidized, the amount of metal remaining was determined chemically by dissolving it in HCl (which does react readily with uranium oxides) and titrating U^{+4} with a standard dichromate solution.

The vertical furnace (Fig. 3.4) required for the continuous recording thermo-balance, comprised a "Mullite" tube 1-1/4" O.D. and 20" long with Pyrex fittings fused to the ends. The apparatus in Fig. 3.4 is the arrangement used for oxidation of uranium in steam.

In most of the experiments conducted in this apparatus, the samples were contained in a 2-1/4" long alumina extraction thimble and were allowed to oxidize completely except in the steam experiments. This is in contrast to those conducted in the open boat and horizontal tube furnace. In practice, it was found that an air flow rate of 300 cc/min or velocity of 120 cm/min (STP) in the horizontal furnace produced an oxidation rate equal to or greater than that produced by 600 cc/min in the vertical furnace with the specimen contained in the deep thimble. Since the two systems differed so widely the amount of surface of metal or oxide directly exposed to the flowing furnace atmosphere, it is not surprising that some differences in results were encountered.

4.1.1 Oxidation of Uranium in Air

Studies of oxidation rates of uranium were reported[21] along with the results of fission-product release determinations.

Effect of Surface Area to Weight Ratio. - Studies made with small specimens showed that even a small variation in surface-to-weight ratio produced a significant change in oxidation rate. These experiments were performed in apparatus of the type shown in Fig. 3.3. Results obtained with an air temperature of 630°C are shown in Fig. 4.1 while results of experiments at 1000 and at 1200°C are displayed in Fig. 4.2.

Effect of Furnace Temperature. - Data on the variation of oxidation rate with furnace temperature covering the range 600 to 1400°C are shown in Fig. 4.3. These data, obtained by use of the continuously weighing balance with an air velocity of 220 cm/min, indicate that the oxidation process is rather complicated, especially in the 600 to 900°C range, and that the rate of oxidation at each temperature varies over a wide range. Possible explanations of the observed oxidation behavior are considered below in the discussion on oxidation of uranium in CO_2.

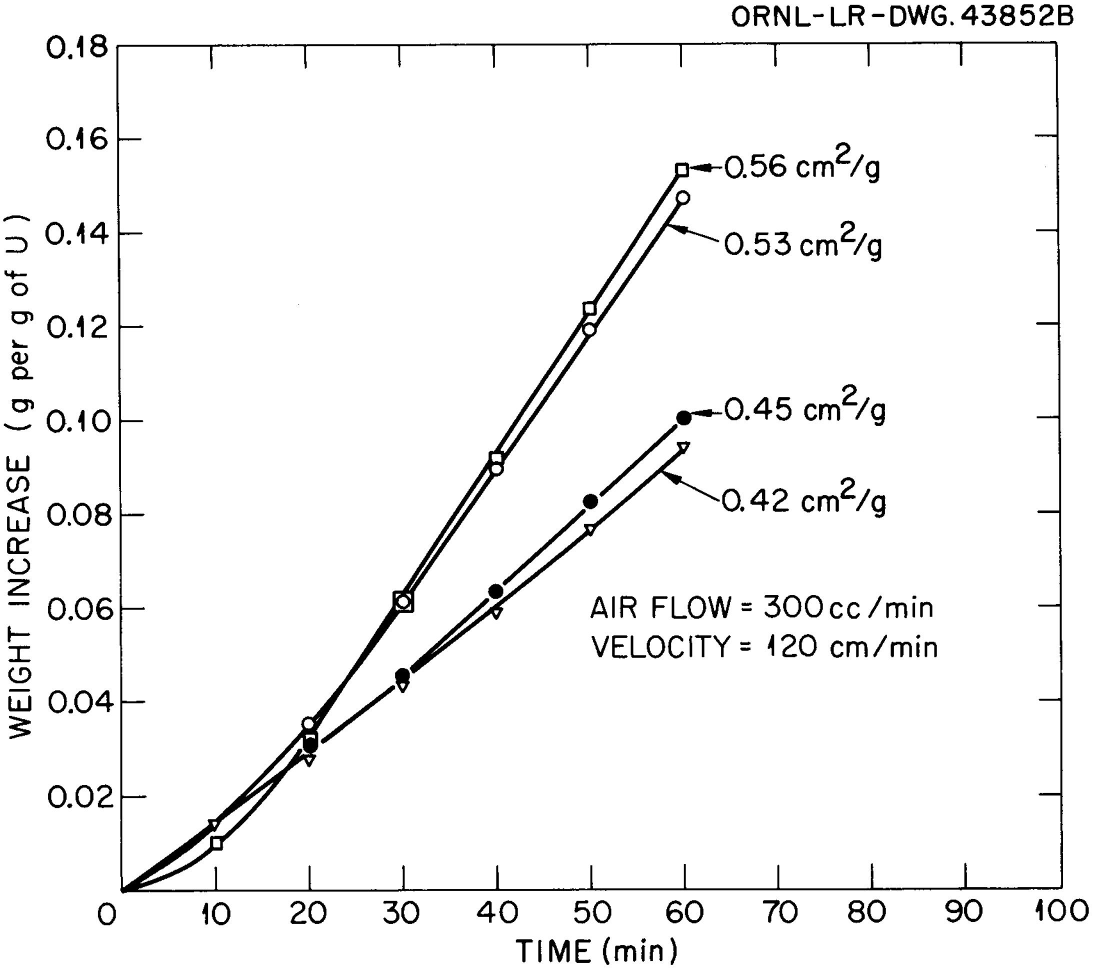

Fig. 4.1. Effect of Surface Area to Weight Ratio on Oxidation of Uranium in Air at 630°C.

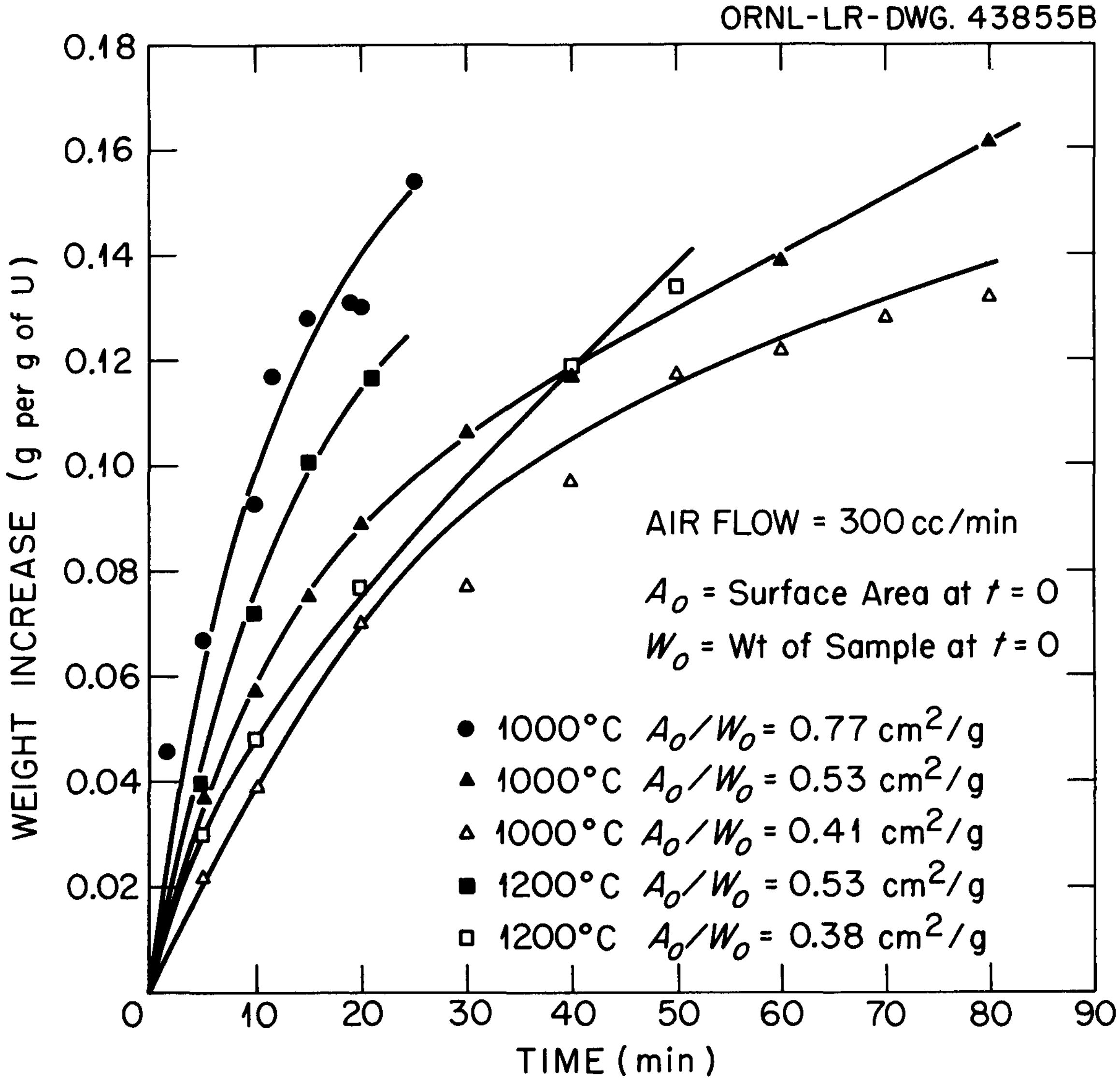

Fig. 4.2. Effect of Surface Area on Oxidation Rate of Uranium in Air.

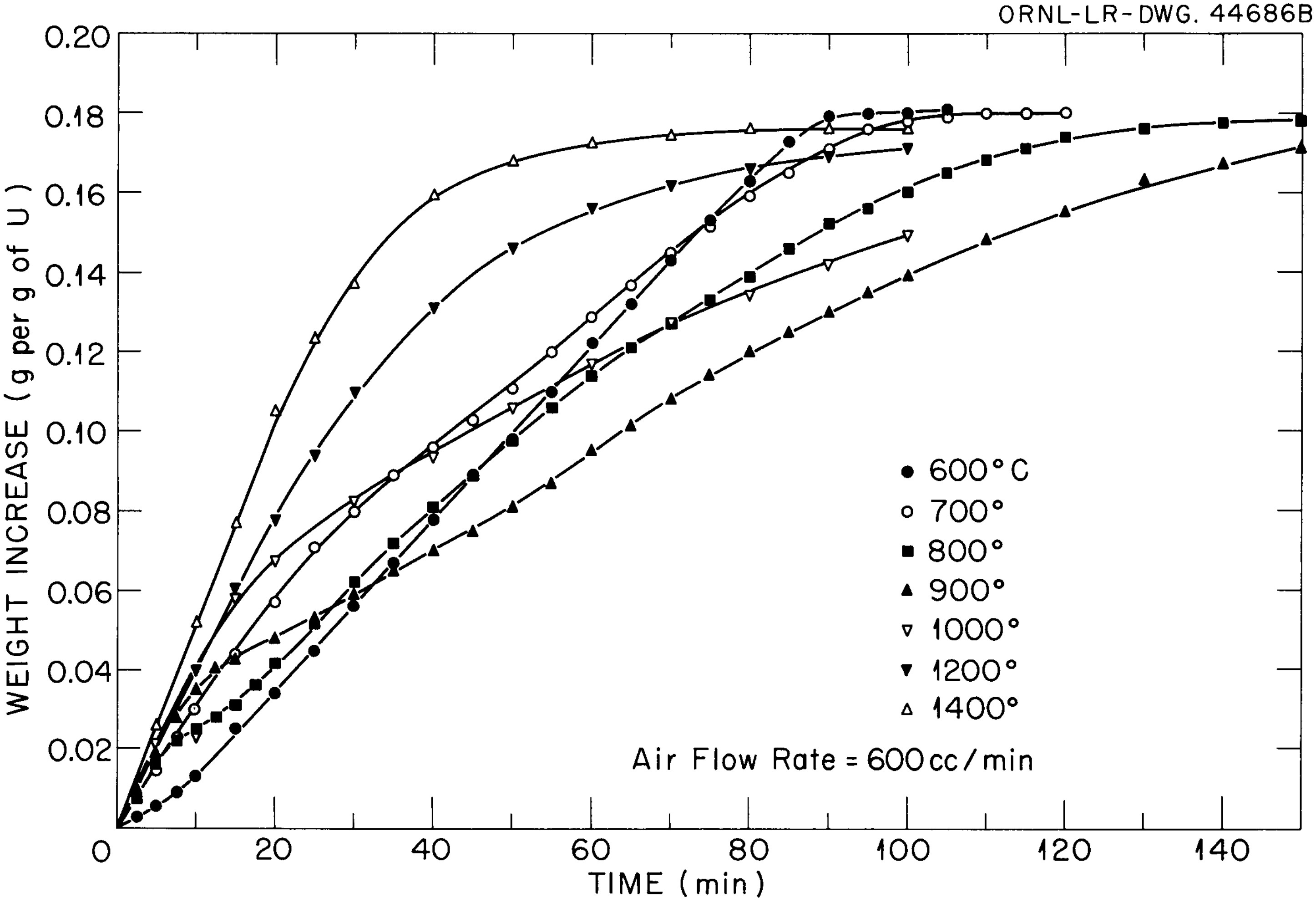

Fig. 4.3. Oxidation of Uranium in Air at Various Temperatures.

Self-Heating Effects. - In most of the uranium oxidation studies, the furnace or furnace gas temperature was measured. The heat of oxidation of uranium is quite large (259 and 285 kcal/gm-atm of uranium for UO_2 and U_3O_8 formation,[29] respectively) and the oxide layer formed serves as an effective heat barrier permitting the temperature of the unoxidized metal to rise considerably above that of the oxidizing atmosphere. It should be recognized that uranium temperatures in much of the published work on oxidation of uranium were higher than furnace temperatures at times because of the self-heating effect.

The extent of self heating of unirradiated uranium resulting from its oxidation in air and oxygen was determined at 1100, 1200 and 1400°C in an experimental arrangement comparable to that in which the partial-oxidation release experiments were carried out (Fig. 3.3). A thermocouple junction was placed in a hole in the specimen and both gas and sample temperatures were continually recorded during heating in helium and during admission of a measured flow of air or oxygen. An optical pyrometer was used to observe the temperature transients beyond the range of the platinum-rhodium thermocouple. Typical temperature rises observed are shown in Table 4.1.

Effect of Burnup. - The oxidation rate of irradiated uranium (~ 0.1% burnup) was compared with that of similar specimens of un-irradiated uranium under the same experimental conditions[21] in the vertical furnace apparatus (Fig. 3.4). (Uranium cylinders, 0.25 in. diameter x 0.7 in. contained in porous alumina thimbles. The gas flow velocity was 220 cm/min measured at room temperature and without allowance for the cross-sectional area of the alumina cup.) Results obtained at gas temperatures of 800, 1000, and 1200°C are shown in Figs. 4.4, 4.5, and 4.6, along with a similar comparison of the effects of irradiation on oxidation rates in CO_2 and steam

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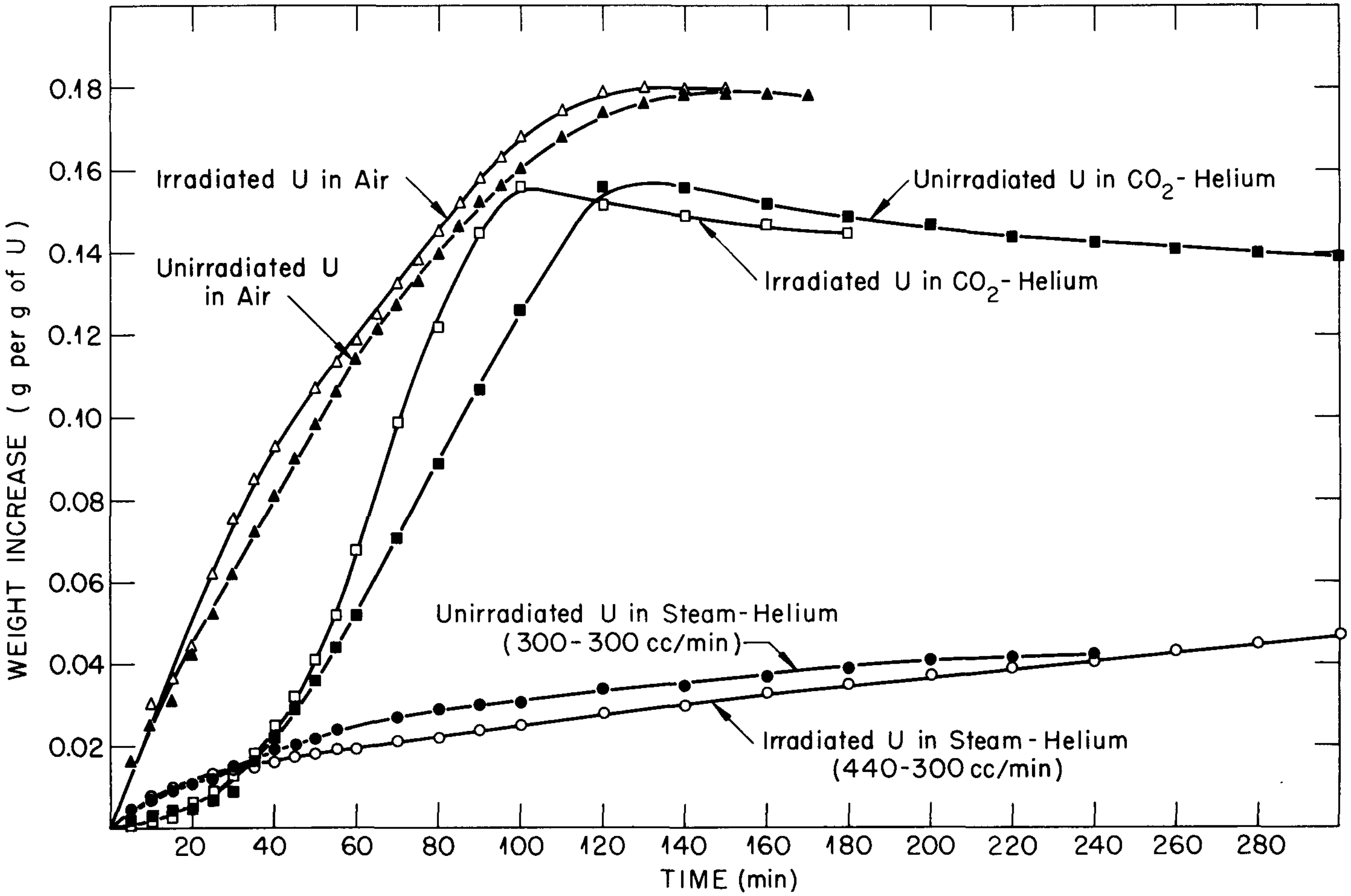

Fig. 4.4. Comparison of Oxidation Rates of Irradiated and Unirradiated Uranium in Air (600 cc/min), CO_2-Helium Mixture (150-450 cc/min) and Steam-Helium Mixture at 800°C.

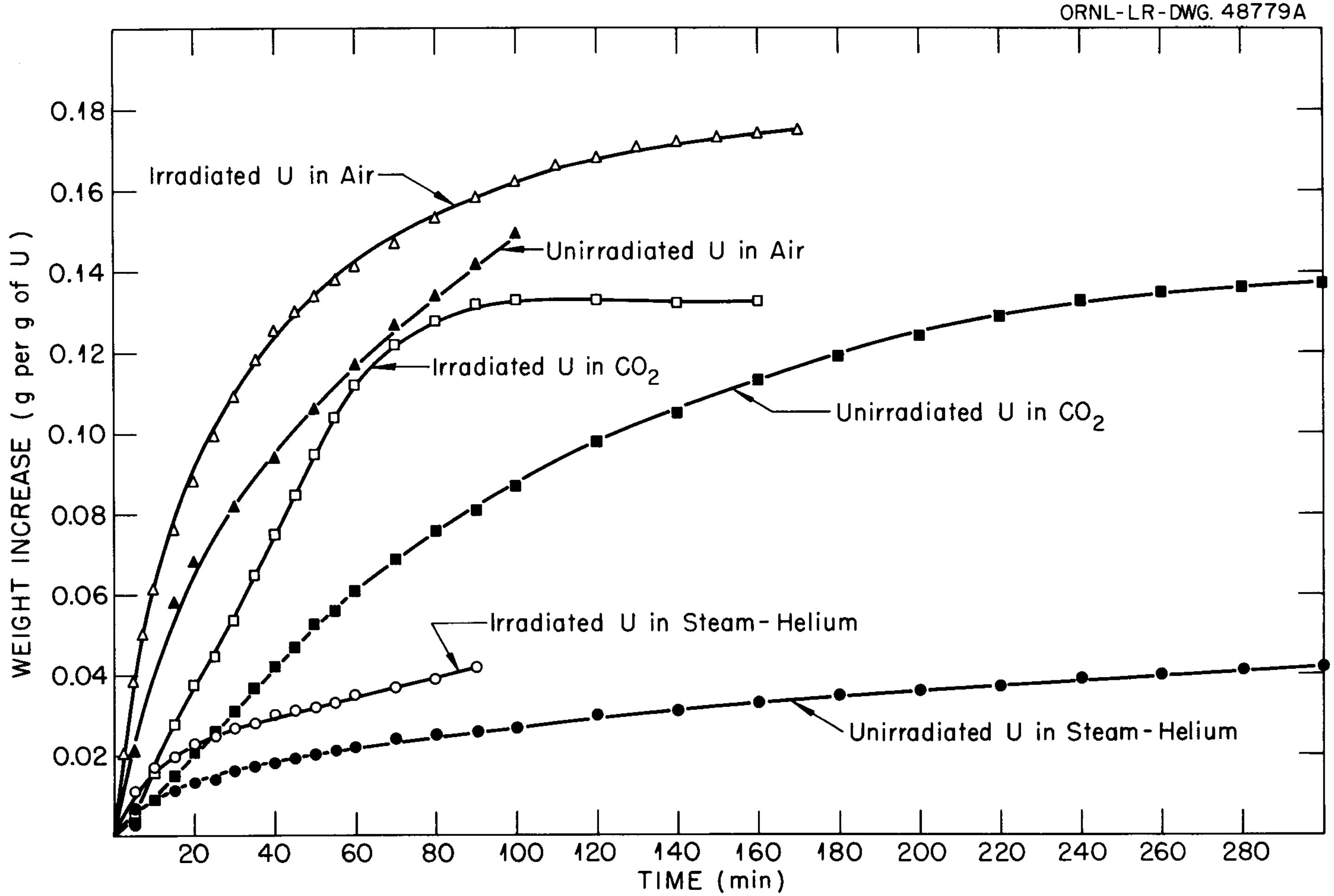

Fig. 4.5. Comparison of Oxidation Rates of Irradiated and Unirradiated Uranium in Steam, Air, and CO_2 at 1000°C. (600 cc/min flow rate).

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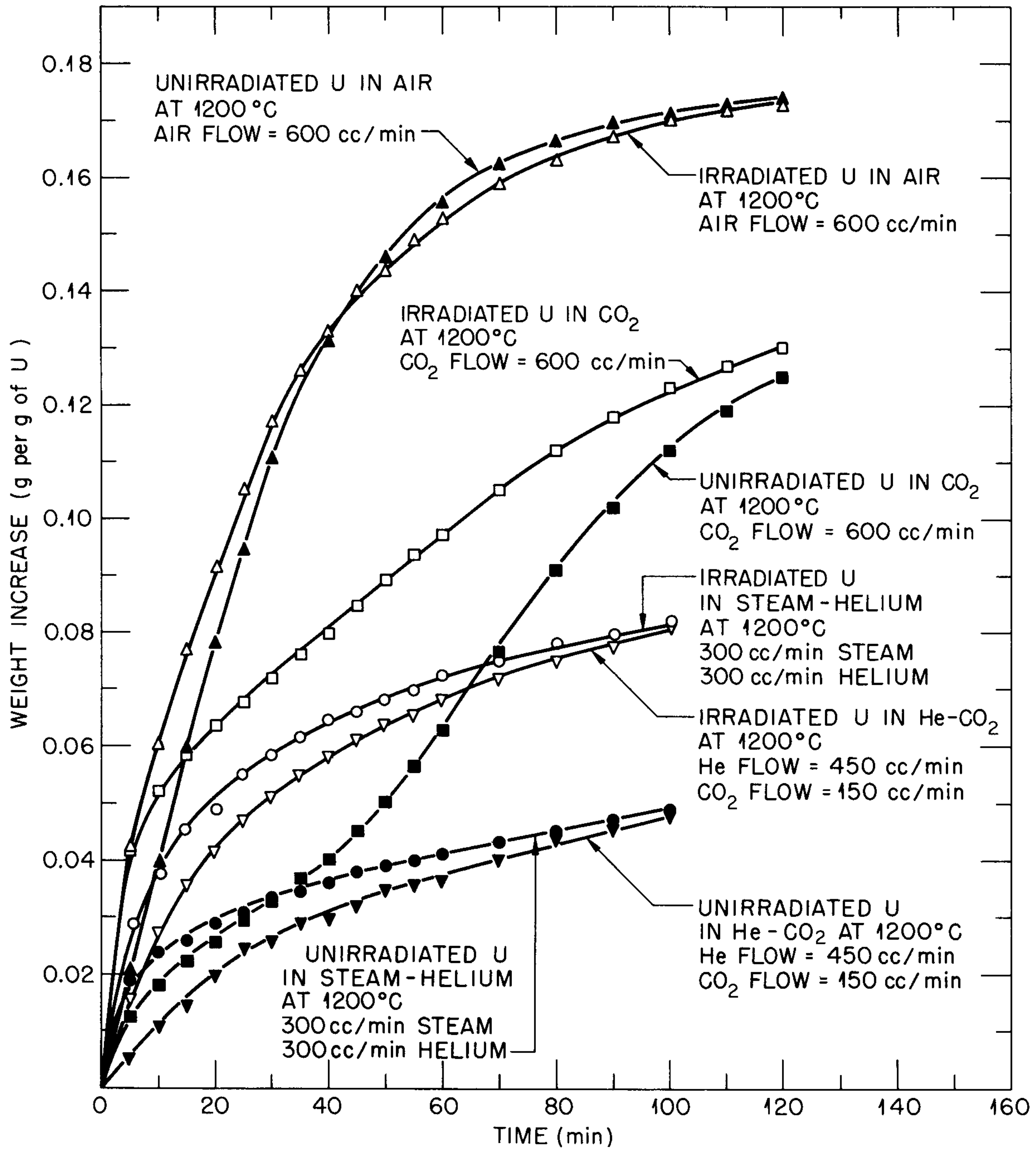

Fig. 4.6. Comparison of Oxidation Rates of Irradiated and Unirradiated Uranium in Steam, Air, and CO_2 at 1200°C.

Table 4.1. Self Heating of Uranium by Heat of Reaction in Air and Oxygen

Sample No.	Initial Temp. of Uranium °C	Max. Temp. Observed During Oxidation, °C		
		400 cc/min Air	300- cc/min O_2	3000 cc/min O_2
1	1100	1250	-	-
2	1200	1375	-	-
3	1200	-	1450**	-
4	1400	1575*	-	-
5	1400	-	-	2650**

* Smoke pattern of U_3O_8 obtained on the filter.

** Heavy plate-out of U_3O_8 obtained on filters.

which will be discussed in subsequent sections. Similar data obtained in the horizontal furnace apparatus (Fig. 3.3) are shown in Figs. 4.7 and 4.8. The data show that irradiated uranium oxidized more rapidly, at least initially, than unirradiated uranium at all three temperatures but the difference was most pronounced at 1000°C. The burnup effect on oxidation rate was also studied by Hilliard and Reid[32] over a broad burnup range at temperatures of 1000, 1200, and 1440°C. More data on this effect would be desirable at higher burnup levels, particularly with larger specimens that would permit extrapolation to surface-to-volume ratios that exist in full-size fuel rods.

4.1.2 Oxidation of Uranium in Carbon Dioxide

Interest in oxidation rates of uranium in CO_2 results from the use of this gas as the coolant in reactors of the Calder Hall type and the possible use of this gas for extinguishing uranium fires.

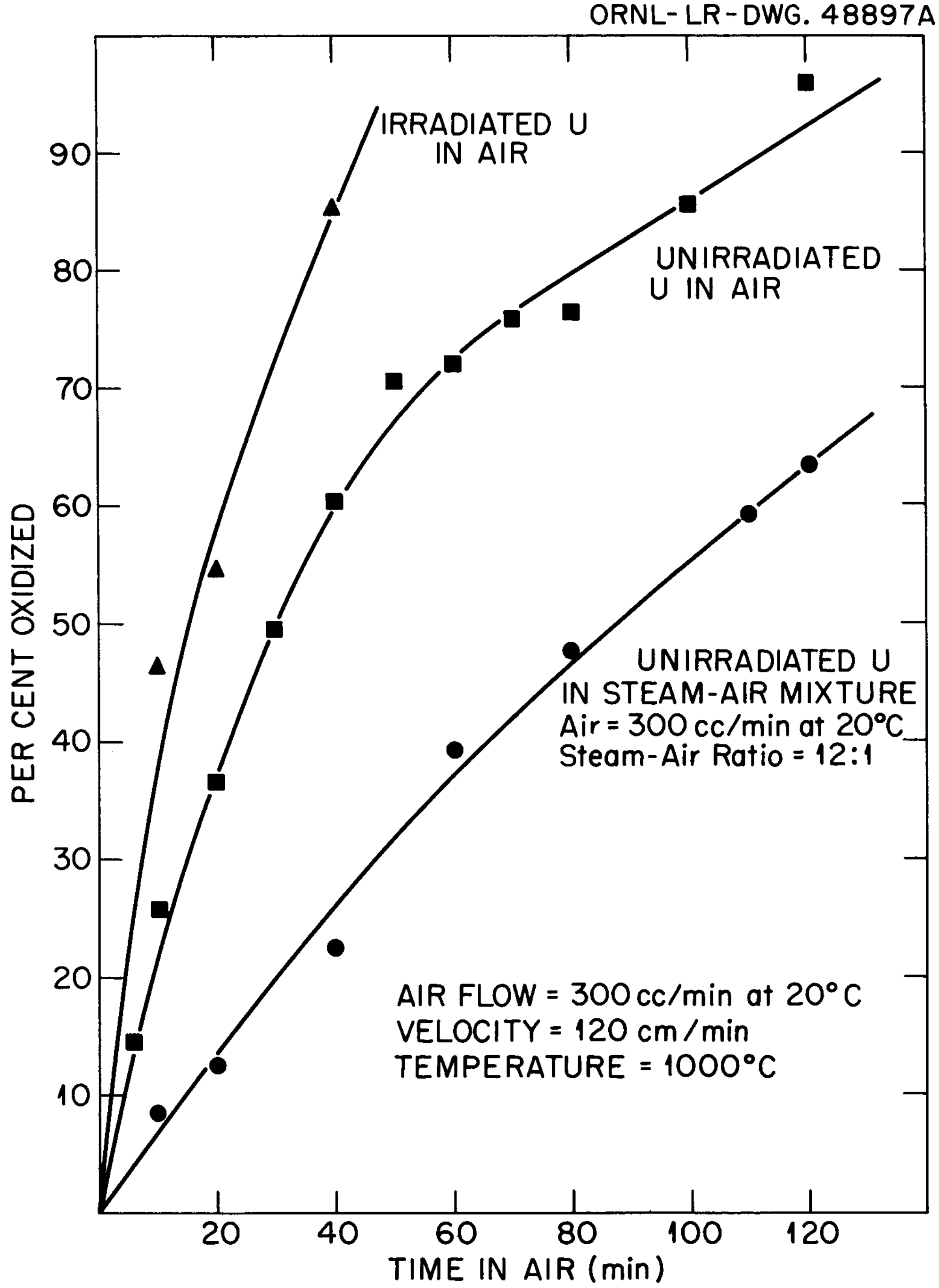

Fig. 4.7. Relative Rate of Oxidation of 1/4-in. x 11/16-in. Cylinder of Irradiated and Non-Irradiated Uranium in Air and Steam Mixture at 1000°C.

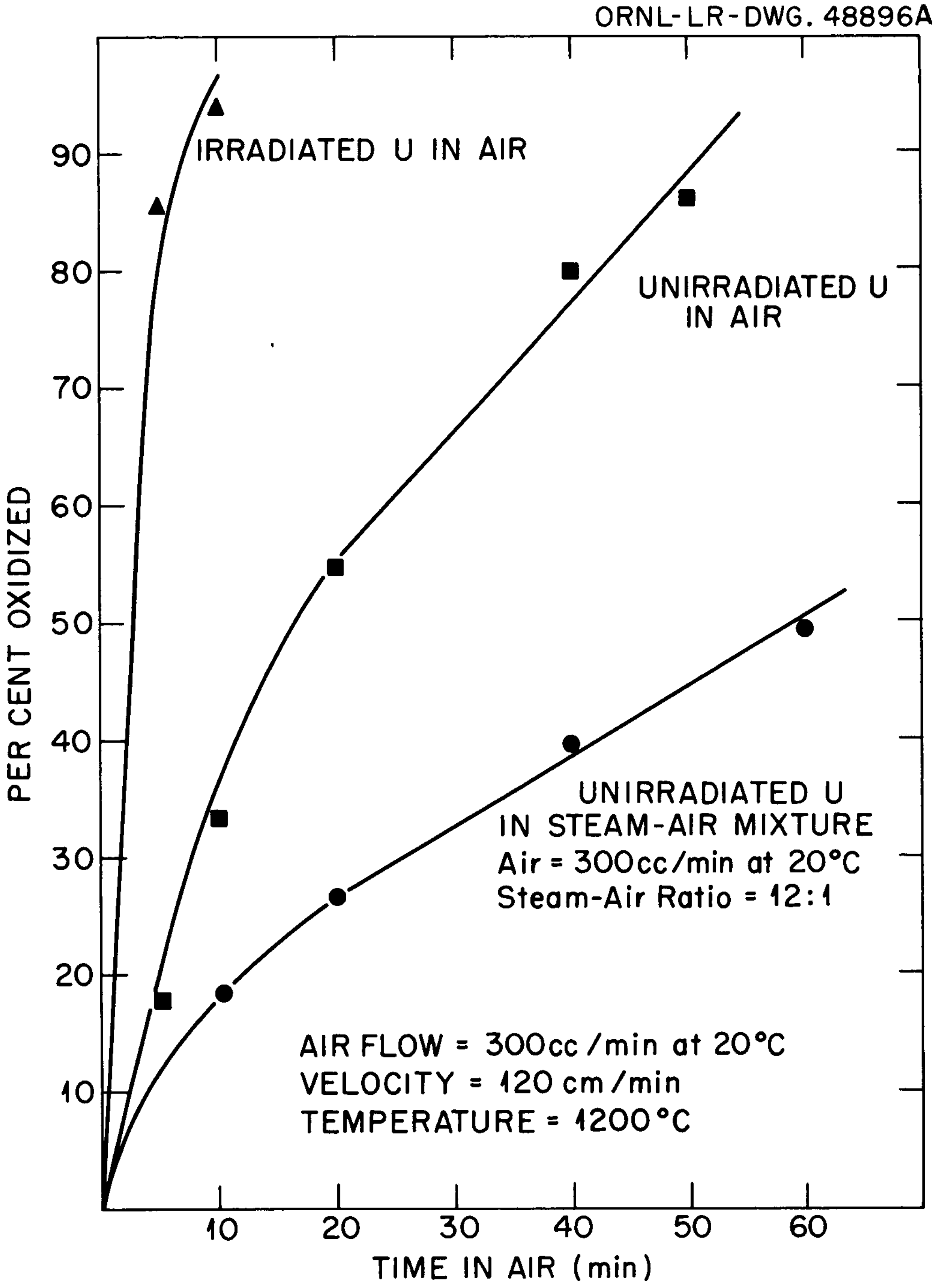

Fig. 4.8. Relative Rate of Oxidation of 1/4-in. x 11/16-in. Cylinder of Irradiated and Non-Irradiated Uranium in Air and Steam Mixture at 1200°C.

Effect of Temperature. - The interesting oxidation behavior of uranium in CO_2 at 800°C is shown in Fig. 4.4. It appears to be due to formation of a metastable oxide during rapid oxidation. The maximum O:U ratio observed was approximately 2.32. Comparison of data shown in Fig. 4.4 and Fig. 4.6 indicates that complete oxidation is not achieved as rapidly at 1200 as at 800°C in spite of the fact that the initial oxidation rate was higher at the higher temperature.

Effect of Gas Flow Rate. - Data obtained by exposure of uranium to undiluted commercial CO_2 at different temperatures and flow rates, Fig. 4.9, showed that the oxidation rate increased with increasing flow rate at 1200°C but not at 1400°C. It is probable that the nature of the protective oxide coating is more important than the gas flow rate.

Effect of Burnup. - The effect of irradiation of uranium on its oxidation rate in CO_2, shown in Figs. 4.4, 4.5, and 4.6 demonstrate that the burnup effect is more pronounced in CO_2 than in air. This effect was also investigated by Diffey and King[47] at irradiation levels of 1250 to 2350 Mwd/T.

Several investigators have discussed possible reasons for the increased oxidation rate of irradiated uranium and the increased release of fission products at high burnups discussed elsewhere in this document. Hilliard and Reid[32] ascribe the increased oxidation rate at temperatures above the melting point of uranium (1133°C) under their experimental conditions to formation of fission gas bubbles which burst through the thin oxide layer covering the molten uranium allowing it to flow and cover the bottom of the crucible. Diffey and King[47] expressed the belief that the very high oxidation rates of irradiated uranium in CO_2 at 800°C were due to swelling caused by release of fission-product gas within the uranium. Buddery and Scott[45] studied bubble formation accompanying the melting of irradiated uranium in some detail and it seems reasonable to assume that increased access of

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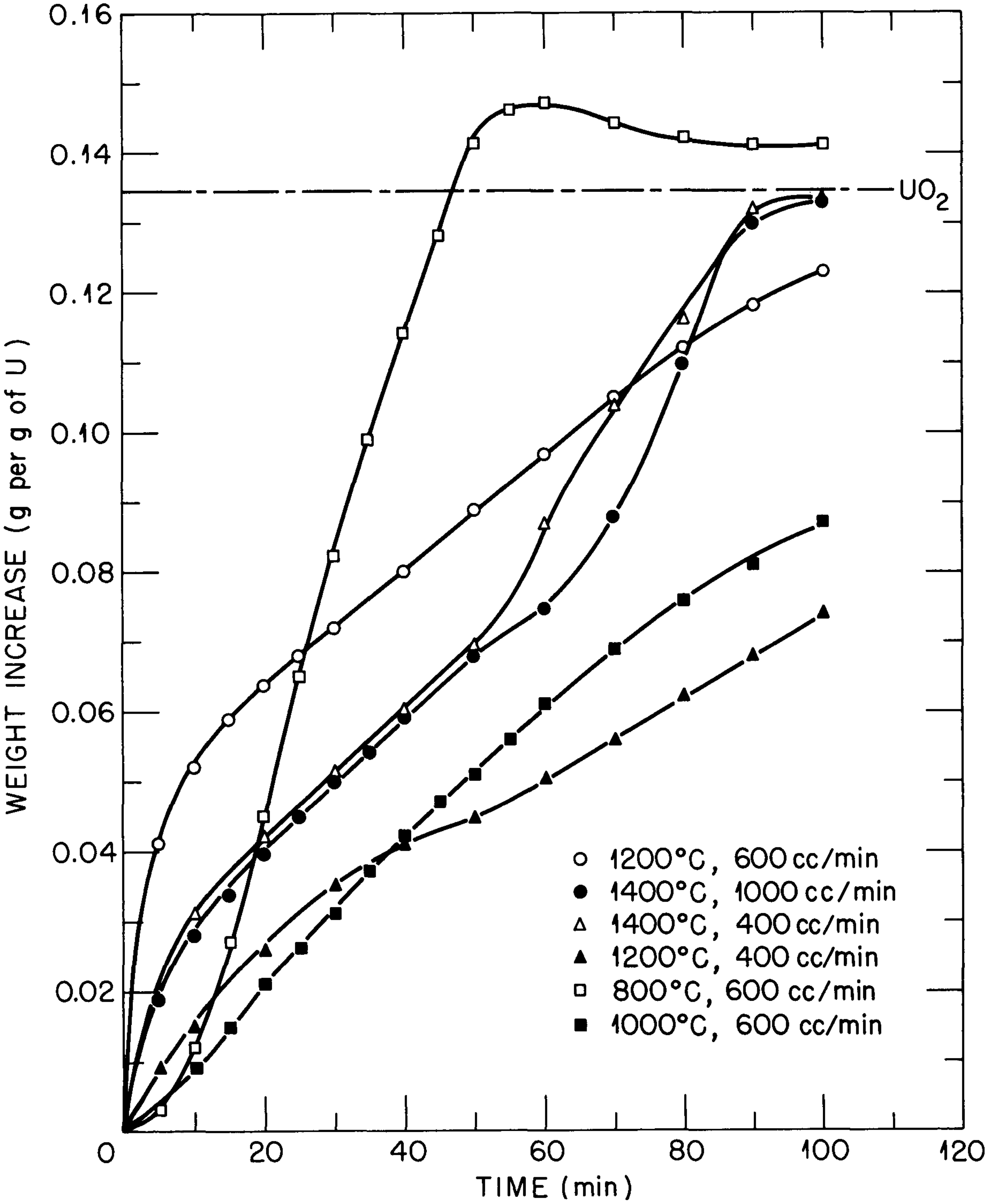

Fig. 4.9. Oxidation of Uranium in CO_2.

oxygen or CO_2 to unoxidized uranium could result from cracks or holes in the oxide coating produced by fission gas. It has also been suggested[21] that imperfections in the oxide coating of irradiated uranium could result from the presence of fission-product atoms and that these imperfections could lead to cracking and diminished protectivity of the oxide coating. Experimental evidence supporting this explanation is lacking at the present time and further study appears to be required in order to provide a more adequate explanation of the burnup effect.

4.1.3 Oxidation of Uranium in Steam

Effect of Temperature and Steam Flow Rate. - Weight-increase data obtained by exposing uranium specimens to steam-helium mixtures at different temperatures are included in Figs. 4.4, 4.6, and in 4.10. The oxidation rate increased, in general, with increasing temperature in the 800 to 1400°C range. It may seem a bit surprising, however, that the oxidation rate shown in Fig. 4.10 decreased with increasing steam flow rate. This result was explained[21] on the basis that the sintering action of steam on UO_2 reported in the literature occurs at a rate proportional to the steam-flow rate.

Inspection of the curves in Figs. 4.4, 4.5, and 4.6 show that the initial oxidation rate was the maximum rate observed. After the initial rapid stage of oxidation was completed, the rate became essentially constant for some time and then gradually decreased if oxidation proceeded for an extended period. The high initial rate is due to the reaction of essentially unprotected uranium with steam.

Effect of Burnup. - A comparison of the oxidation rates of irradiated and unirradiated uranium in steam at 800 to 1000°C, shown in Figs. 4.4 and 4.5, show the same type behavior that was observed in the tests made in air and in CO_2. The irradiation effect was not observed in the tests made at

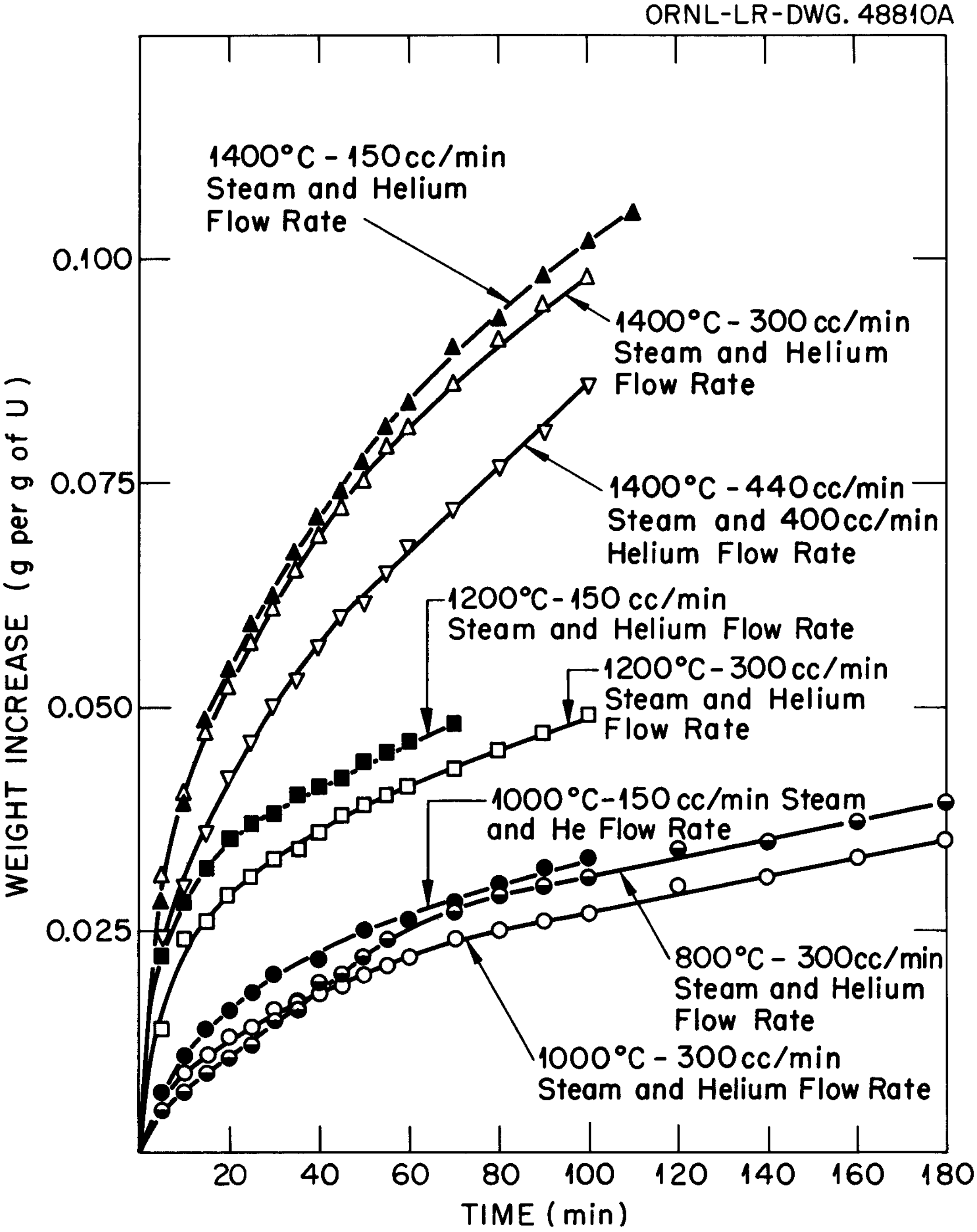

Fig. 4.10. Effect of Varying Temperature and Steam Flow Rate on Oxidation of Uranium in Steam Diluted with Helium.

$1200^{\circ}C$ (Fig. 4.6) because of the higher steam flow employed with the irradiated specimens. As noted above, increased steam-flow rates produce lower oxidation rates in steam.

4.1.4 Oxidation in Steam-Air Mixtures

The scanty information that was obtained on oxidation of uranium in a steam-air mixture (12:1 volume ratio) is displayed in Figs. 4.7, and 4.8, along with data on oxidation of irradiated and unirradiated uranium obtained under comparable conditions in the horizontal furnace apparatus (Fig. 3.3). These data show that the oxidation rate in air was much faster than in an atmosphere that was largely steam.

4.1.5 Comparison of Oxidation Rates in Various Atmospheres

The data shown in Figs. 4.4, 4.5, and 4.6 permit ready comparison of the oxidation rates of both irradiated and unirradiated uranium in three different atmospheres. The rates are in the same decreasing order, air, CO_2 and steam at all three temperatures used in these experiments. It is observed that complete oxidation of irradiated uranium occurred about as fast in CO_2 as in air at 800 and $1000^{\circ}C$ while none of the specimens were completely oxidized in steam at these temperatures. These data would have permitted prediction of the observed ineffectiveness of CO_2 for quenching the Windscale Pile No. 1 fire if they had been available at that time. The data also show that steam provides the minimum oxidation rate and, consequently, the successful use of water in quenching the Windscale fire could also have been predicted.

4.2 Release of Fission Products from Metallic Uranium

Data included in this section were reported in Ref. 21.

4.2.1 Uranium Melted in an Inert Gas

Studies of the release of fission products accompanying the melting of irradiated uranium were carried out at Ames[1] and at the Argonne National Laboratory. Burris et al.[2] reported volatilization of a large fraction of the rare gases, halogens, and cesium. Hilliard[20] heated 11.5-gm cylinders of tracer-level-irradiated uranium in helium and in air and gave a comparison of fission product release.

Several experiments involving the melting of irradiated uranium (0.1 atom % burnup) in impure helium were performed. The data obtained are given in Table 4.2. The lack of correlation between the amount of uranium oxidized and the fraction of fission gases released indicates that oxidation had a minor effect on release in these experiments. The discrepancy between the high gas release values and low value reported in early studies by another investigator[20] can probably be attributed to the difference in gas concentration in the uranium used in the two investigations (approximately 10^6 greater concentration of xenon in the ORNL experiments). Later Hanford data[32] obtained with higher burnup fuel are more nearly in agreement with ORNL results.

Table 4.2 Fraction of Rare Gases Released on Melting Irradiated Uranium (0.1 atm % burnup) in Impure Helium

Maximum Furnace Temperature (°C)	Percent of Uranium Oxidized	Percent of Rare Gases Released
1200	4.1	97.7
1170	0.9	98.9
1180	3.9	97.1
1250	15.3	99.6

In some experiments a sensitive in-stream γ-detector ahead of the cold charcoal trap permitted observation of fission-gas release in more detail than was afforded by the use of traps alone. The data shown in Fig. 4.11 indicated that a large fraction of the fission gases was released very rapidly when the uranium melted. The fraction released, as shown by trapping the released gas in refrigerated charcoal, only increased from 98% in 16 minutes after melting began to 99.3% after 38 minutes. A sharp peak in the counter trace that recorded the rate of gas release during the cooling period was noted at the freezing point of uranium. A smaller peak was noted that probably corresponded to the β-α inversion point (660°C) and another large peak was noted when the uranium was cooled rapidly from 513°C to room temperature. These observations indicate that a part of the fission gas not released while irradiated uranium is in the molten condition will be "squeezed" out during the cooling period but the fraction released by this process is probably too small to be of significance in hazards analyses.

4.2.2 Uranium Oxidized in Air

Data on fission-product release from irradiated uranium (0.1 atom % burnup), partially oxidized in the horizontal furnace-tube apparatus (Fig. 3.3) are shown in Table 4.3. The cylindrical specimens, with a diameter of 0.25 in. and an approximate length of 0.75 in., were contained in shallow boats inside a quartz furnace tube. They were heated and cooled in flowing helium and exposed to air for varying lengths of time at two furnace temperatures. The data confirm the results of Hilliard's[20] studies made with similar specimens irradiated to 2.4×10^{14} nvt which showed increasing release of the more volatile fission products with increasing fraction of uranium oxidized. However, the release of iodine and tellurium from the low-burnup uranium appeared to level out at about 80% in his experiments whereas the release of these fission products from the oxidized portion of higher burnup material, as shown in Table 4.3, appeared to be

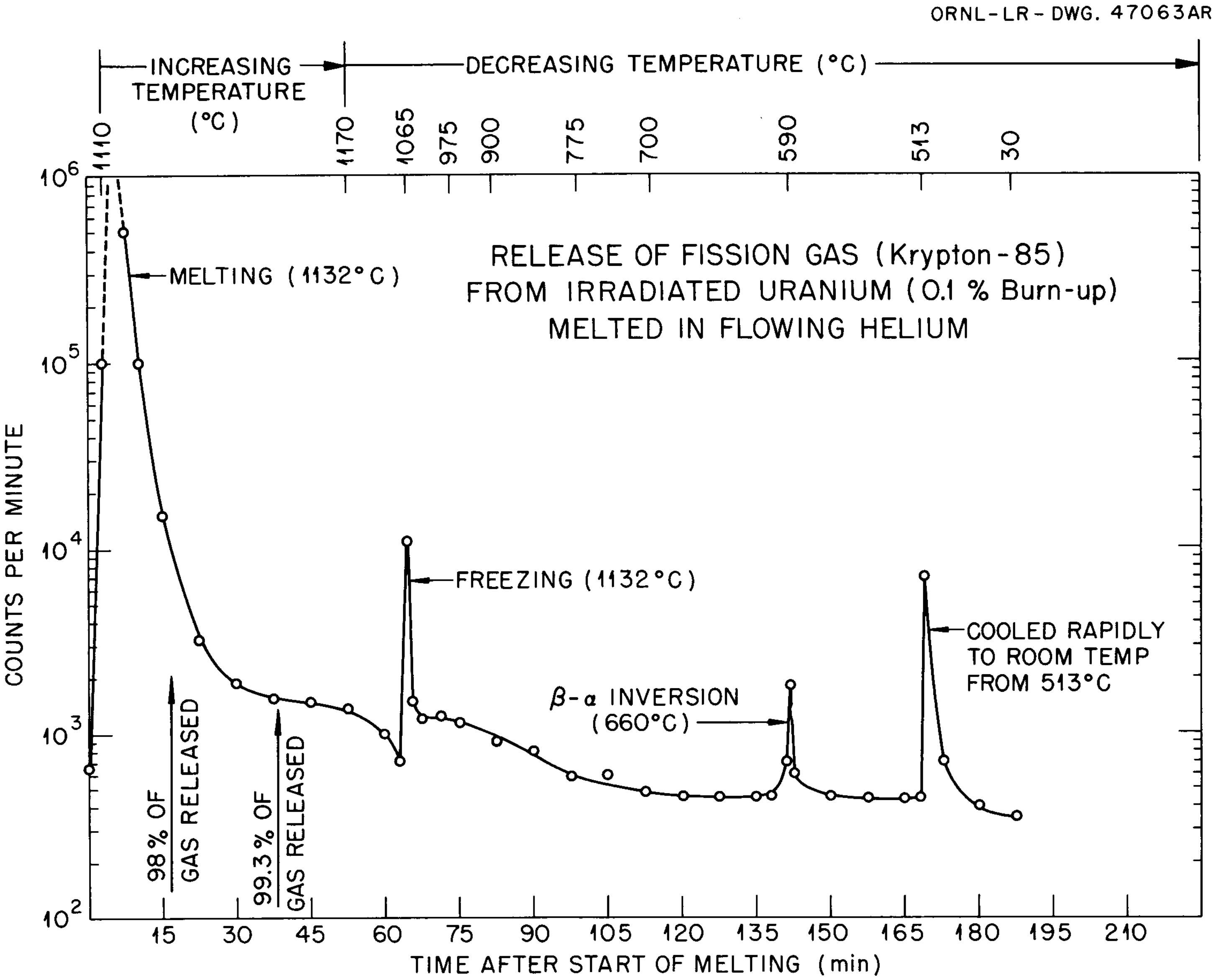

Fig. 4.11. Release of Fission Gas (Krypton-85) from Irradiated Uranium (0.1% Burnup) Melted in Flowing Helium.

Table 4.3 Fission-Product Release From Irradiated Uranium[a] Incompletely Oxidized in Air[b]

Furnace Temp. °C	Time Min.	Percent U Oxidized	Percent of Total Activity Released							
			Xe-Kr	I	Te	Cs	Ru	Zr	Ce	Sr
1000	<1	11.1	-	3.1	-	0.01	0.004	0.003		
1000	10	46.9	~100	-	-	2.4	0.1	0.007	0.001	0.05
1000	20	53.2	~100	67.3	-	2.8	0.13	0.005	0.008	0.055
1000	40	86.9	~100	79.5	-	18.4	5.2	0.018	0.006	0.05
1200	<1	25.0	-	12.5	-	1.62	0.019	0.17		
1200	5	43.6	97.7	31.9	8.1	18.5	0.035	-	-	0.024
1200	8	66.2	99.2	23.3	12.7	14.5	0.16	-	-	0.028
1200	10	94.0	100			11.2	0.51	0.05	0.06	2.7
1200	10	68.0	98.7	39.9	24.8	17.1	0.22	-	-	0.6
1200	12	77.5	99.8	46.4	23.0	-	-	-	-	0.9
1200	15	72.0	99.8	52.8	51.6	28.6	4.3	-	-	3.1
1200	15	64.8	99.6	71.5	68.0	13.65	2.0	-	-	1.1
1200	20	65.4	99.4	57.3	71.3	13.0	1.8	-	-	0.85
1200	30	72.3	-	62.1	77.4	19.2	2.34	-	-	1.77

[a] 0.1% burnup, pre-heated in helium.

[b] Velocity 120 cm/min, measured at room temperature.

essentially complete. In general, the release of tellurium was somewhat lower than that of the iodine and less cesium was released than tellurium. Moderate releases of ruthenium and strontium were noted at 1200°C, but very little of the cerium and zirconium volatilized.

Data on the distribution of fission products liberated by the complete oxidation of irradiated uranium (0.1 atom % burnup) in an air stream are shown in Table 4.4. These data, which were obtained by use of the vertical furnace tube apparatus (Fig. 3.4), show that release of iodine and ruthenium was quite high even with a furnace temperature of 800°C while the release of cesium and tellurium was rather low. Increasing the furnace temperature to 1000°C produced a moderate increase in cesium release but a large increase in the fraction of iodine and tellurium released. At 1200°C, the release of iodine, tellurium, and ruthenium was essentially complete and a substantial fraction of the cesium also escaped. Very small amounts of the refractory elements cerium, zirconium, and strontium were released.

Iodine was found mostly in the hot (200°C) charcoal bed where it would be expected to be if it was liberated in the molecular form. Most of the cesium remained in the mullite furnace tube, possibly in the form of Cs_2O. The distribution of tellurium varied considerably in the four experiments but it appears that a large fraction of tellurium released by uranium oxidizing in air will be in the form of particulate matter, probably TeO_2. Ruthenium was undoubtedly released as a volatile oxide, RuO_3 or RuO_4, but it probably was quickly converted to a less-volatile lower oxide which accounts for the fact that a large fraction of this element remained in the furnace tube. The part that was airborne long enough to reach the filters stopped there.

Table 4.4. Activity Released by Complete Oxidation of Irradiated Uranium[a] in Air[b]

Experiment Number	Temp. °C	Xe-Kr	Percent of Activity Released						
			I	Cs	Ce	Te	Ru	Zr	Sr[c]
68	800		48	0.06	0.001	2.9	73	0.05	0.002
81	1000	97.1	89	0.4	0.002	80	77	0.02	0.002
83	1200	99.2	90	14	0.0006	96	85	0.01	0.02
32	1200		99.2	16	0.03	84	78	0.08	0.005

[a] 0.1 atom % burnup.

[b] Air flow velocity, 220 cm/min measured at room temperature.

[c] Values probably low due to chemisorption of SrO by the mullite furnace tube.

4.2.3 Uranium Oxidized in CO_2

There is comparatively little information in the literature on the release of fission products from uranium oxidized in CO_2. Complete oxidation of irradiated uranium (0.1% burnup) at 800, 1000, and 1200°C in commercial grade CO_2 (1 to 2% O_2) or in CO_2 diluted with helium gave release data recorded in Table 4.5. The data show that cesium and ruthenium release values obtained in CO_2 were very much lower than the corresponding values obtained in air (Table 4.4) except for the anomalous ruthenium release result in Experiment 84, discussed below. Iodine and tellurium were released to about the same extent in CO_2 that they were in air at the same temperature.

In Experiment No. 84, Table 4.5, a very large fraction of the ruthenium was released under unusual circumstances. Approximately 10% of the uranium was inadvertently oxidized in air at the beginning of the experiment and, after oxidizing the remaining uranium in CO_2, a high-velocity stream of helium was passed through the apparatus for 30 minutes. This combination of atmospheric conditions produced higher release values of several nuclides than resulted from oxidation in CO_2 at the same temperature under normal conditions (Experiment No. 31), but the behavior of ruthenium in this experiment is especially noteworthy. All the filter papers used to collect particulate matter in this experiment contained ruthenium, including, in order, a 3.0μ Millipore, a 0.4μ Millipore, a coarse cellulose Whatman paper, a 0.4μ Millipore, and another Whatman cellulose paper. This fact indicates that the ruthenium was in the form of very small particles or a gas and suggests that ruthenium probably volatilized as RuO_3 or RuO_4 for reasons that are not clear. The existence of released ruthenium in the form of an oxide was suggested by the fact that it dissolved easily. Ruthenium oxide is much easier to dissolve than the metal.

Table 4.5. Fission-Products Released by Complete Oxidation of Irradiated Uranium[a] in CO_2[b] or in CO_2 Diluted with Helium[c]

Experiment No.	Atmosphere	Temp. °C	Gross γ	Percent of Total Activity Released							Rare Gases
				I	Cs	Ce	Te	Ru	Zr	Sr	
67	CO_2-He	800	0.7	5.8	0.02	0.001	1.9	0.04	0.044		Not determined
82	CO_2	1000	0.7	85	0.002	0.0003	39	0.08	0.002	0.001	75
62	CO_2-He	1200	0.6	68	0.9	0.003	95		0.59	0.012	Not determined
31	CO_2	1200	7.6	53	1.7	0.002	69	2.0	0.09	0.01	Not determined
84	CO_2[d]	1200	14.6	85	1.8	0.004	96	93[e]	0.020	0.3	99.2

[a] 0.1 atom % burnup.

[b] CO_2 flow velocity, 200 cm/min, measured at 25°C.

[c] CO_2 flow rate, 150 cc/min; helium flow rate, 450 cc/min.

[d] Approximately 10% of the uranium was oxidized by accidental admission of air at the beginning of this run. After the completion of the CO_2 oxidation, high-velocity helium (125 fpm) was passed through the apparatus for 30 min.

[e] Average of two analyses. High results probably due to unusual conditions mentioned above.

4.2.4 Uranium Oxidized in Steam-Helium Mixtures

The principal published reports on the release of fission products from irradiated uranium oxidized in steam are those of Scott[49] and of Parker et al.[21]. The latter investigators diluted their steam with helium and results of these studies are summarized in this section.

Experiments on the release of fission products from irradiated uranium (0.1% burnup) oxidized in steam were most conveniently performed through use of helium as the carrier gas. In addition to serving as an inert carrier of steam, the helium swept fission gases into the cold charcoal trap after steam was removed from the furnace exit gas mixture by condensation. Release data obtained when irradiated uranium specimens were exposed to a mixture of steam and helium at 800, 1000, and 1200°C are displayed in Table 4.6. The slow oxidation rates attained in this atmosphere made it impractical to oxidize the uranium completely. Consequently, the release values shown in Table 4.6 should be divided by the fraction of uranium oxidized before comparing them with the data in Tables 4.4 and 4.5. This correction ignores loss of fission products through diffusion from unoxidized portions of the specimens which probably accounts for the fact that adjusted values of more volatile elements such as iodine and tellurium exceeded 100% in some cases. The data in Table 4.6 show that the dense, adherent, coating of UO_2 formed around uranium specimens exposed to steam at temperatures in the range 800 to 1200°C resulted in marked reduction of the fraction of fission products released, except for tellurium. These data confirm Scott's values[49] showing that iodine and tellurium are released to approximately the same extent as the rare gases in this environment. The high tellurium release value obtained in steam at 1200°C, as compared with iodine release observed at this temperature, suggests the possibility that hydrogen released by the uranium-steam

Table 4.6. Fission Products Released by Incomplete Oxidation of Irradiated Uranium[a] in Steam Diluted with Helium[b]

Temperature °C	Percent of U Oxidized	Percent of Total Activity Released								
		Gross γ	I	Cs	Ce	Te	Ru	Zr	Sr	Xe-Kr
800	36	0.002	0.3							0.9
1000	34	0.1	5.2	0.04	0.007	2.0	0.02	0.04	0.02	3.0
1200	65	0.2	15	0.2	0.0006	79	0.01	0.2	0.02	

[a] 0.1% burnup, preheated in helium.

[b] Steam and helium flow rate each approximately 300 cc/min, measured or calculated rate at room temperature.

reaction may have combined with tellurium to form highly-volatile H_2Te. However, no corroborating evidence for the formation of such a compound was noted in these experiments and Scott's data, which show the same range of iodine and tellurium release values at 1215°C and higher iodine than tellurium release at 1440°C, do not appear to support this hypothesis. The low cesium, ruthenium, and rare gas-release values observed in a steam atmosphere are especially noteworthy.

4.2.5 Uranium Heated in Steam-Air Mixtures

The only available data on fission-product release from irradiated uranium oxidized in steam mixed with air are contained in Table 4.7. These data were obtained[21] with tracer-level (~ 10^{17} nvt or 10^{-4} atom % burnup) irradiated cylindrical specimens weighing about 11 grams. The steam-to-air ratio by volume was calculated to be 12 to 1 at 20°C and the air flow velocity was 120 cm/min, measured at 20°. Horizontal furnace tube-open boat apparatus was employed in these experiments. The data, when adjusted for fraction of uranium oxidized, are comparable to the air oxidation data shown in Table 4.4, except for the ruthenium results.

4.2.6 Comparison of Fission-Product Release From Irradiated Uranium in Various Atmospheres at 1200°C

Release values obtained on heating irradiated uranium (0.1% burnup, except for one test) in various atmospheres at 1200°C are compared in Table 4.8. The high ruthenium release obtained on completely oxidizing the specimen in air, as compared to partial oxidation in CO_2, is quite noticeable.

4.3 Release of Fission Products from Aluminum-Uranium Alloys

Aluminum-uranium alloys clad with aluminum have been employed extensively in research reactors (LITR and ORR at

Table 4.7. Fission-Product Release from Tracer-Level-Irradiated Uranium[a] Heated in Air-Steam Mixture[b]

Furnace Temp. °C	Time min.	Percent of U Oxidized	Percent of Total Activity Released					
			Xe-Kr	Gross γ	I	Te	Cs	Ru
1000	11	17	4.7	0.5	4.3	1.8	0.8	0.4
1000	40	25	5.5	0.3	4.9	6.2	0.4	0.2
1000	60	42	-	0.3	2.7	4.7	0.2	0.02
1200	10	13	26	8.9	8.6	6.5	1.6	0.04
1200	40	28	65	5.9	46	42	5.8	0.3
1200	61	49	-	4.9	47	60	6.9	4.0

[a] Approximate burnup level 10^{-4} atom %; preheated in helium.

[b] Steam to air ratio by volume was 12:1 at 20°C. Air flow velocity was 120 cm/min, measured at room temperature.

Table 4.8. Comparison of Fission-Product Release from Irradiated Uranium[a] Heated in Various Atmospheres at 1200°C.

Atmosphere	Time Heated (min)	Percent of U Oxidized	Percent of Total Activity Released						
			Xe-Kr	I	Te	Cs	Ru	Sr	Zr
Air	20	65	99.4	57	71	13	1.8	0.09	~0.05
Air	250	100	99.2	90	96	~14	85	-	0.01
CO_2	630	100	~99	53	69	1.7[b]	2.0	0.01[b]	0.1
CO_2-helium	410	90		68	95	0.9[b]		0.01[b]	0.6
Steam-helium	123	65		15	79	0.2[b]	0.01	0.02[b]	0.2
Helium	148	4	98	47	0.6	2.0		0.9	
Air[c]	50	92	81	92	66	~1.0	0.6	0.01	0.007
Steam[c]	120	33	4	18	12				

[a] 0.1 atom % burnup, preheated in helium.

[b] Release values probably low because of chemisorption in mullite furnace tube or alumina crucible.

[c] Tracer-level irradiation.

ORNL) and in materials testing reactors (MTR and ETR at the NRTS). Early data on fission-product release from fuel elements of this type were obtained[8] with low burnup fuel materials. The release of fission products from irradiated U-Al alloy specimens has been recently studied over a wide range of temperatures in several atmospheres, using fuels of several degrees of burnup.[43,50] In most of the recent experiments, the fuel was held at the maximum temperature for two minutes but the specimens were molten for periods ranging from about 10 to 17 minutes due to the time required for heating and cooling. Some experiments were performed with longer heating periods because of the possibility that fission-product decay heat could maintain this low-melting fuel material in the molten state for considerable lengths of time after a loss-of-coolant accident.

Data[50] showing the effect on fission-product release of several variables including temperature, atmosphere, time at temperature, and air flow rate are given in Table 4.9. These data were all obtained with fuel specimens irradiated to 23.6 atom % ^{235}U burnup. It is apparent that, at this burnup level, release of rare gases is almost quantitative at any temperature above the melting point of the fuel. Release of other elements increased, in general, with increasing temperature as might be expected. The effect of atmosphere is most noticeable in the tellurium and cesium results. Release of cesium was much higher than that of tellurium in helium while the order was reversed in air and steam-air atmospheres, at least at high temperatures. The release of iodine was slightly higher in oxidizing atmospheres than in helium and, at 900°C or higher temperatures, more than 90% of this important fission product was released in the presence of oxygen. Mixing steam with air had no significant effect on fission-product release and this is in marked contrast to the effect of steam on fission-product release from metallic uranium. Increasing the time at maximum temperature in air produced a moderate

Table 4.9. Effect of Maximum Temperature, Time at Temperature, and Atmosphere[a] on Fission-Product Release from U-Al Alloy Specimens[b]

Maximum Temp. (^{o}C)	Time at Maximum Temperature (min)	Atmosphere[a]	Release (%)					
			Gross γ	Rare Gases	I	Te	Cs	Ru
800	2	Helium	7.4	99.5	29.8	5.3	13.0	0.18
900	2	Helium	13.5	~100	52.8	4.3	20.8	0.08
1000	2	Helium	23.5	~100	82.1	2.9	47.7	0.19
1105	2	Helium	40.7	~100	82.4	2.9	69.5	0.25
700	2	Air	2.3	97.9	37.8	0.3	3.1	0.02
800	2	Air	3.1	99.4	78.6	0.2	3.8	<0.1
900	2	Air	5.2	100.0	91.9	2.1	6.2	0.1
1000	2	Air	6.7	99.8	97.3	<9.7	8.8	0.2
1090	2	Air	12.0	100.0	98.4	44.8	12.4	0.6
1145	2	Air	16.8	100.0	94.2	62.0	18.6	0.4
700	2	Steam-Air	0.9	98.3	27.0	<0.03	0.6	<0.02
800	2	Steam-Air	2.5	99.5	76.8	0.3	1.1	0.1
900	2	Steam-Air	6.8	99.9	90.6	5.7	6.5	0.5
1000	2	Steam-Air	10.6	~100	95.6	22.6	11.0	0.5
1085	2	Steam-Air	25.5	~100	96.8	67.9	30.5	0.8
700	60	Air	3.3	97.7	58.0	<0.14	3.5	<0.02
800	60	Air	4.5	99.5	84.7	0.7	5.9	0.03

(continued next page)

Table 4.9. (continued)

Maximum Temp. (°C)	Time at Maximum Temperature (min)	Atmosphere[a]	Release (%)					
			Gross γ	Rare Gases	I	Te	Cs	Ru
900	60	Air	6.3	99.95	95.3	2.9	9.2	0.2
1000	60	Air	18.1	99.98	92.8	16.6	23.3	0.
1090	60	Air	16.1	99.98	98.3	78.4	37.8	0.03
840[c]	60	Air	5.3	~100	94.6	1.5	6.5	0.1
870[c]	60	Air	8.1	~100	95.8	4.0	6.9	0.7

[a] Gas flowing at a rate of 250 cc/min (measured at room temperature) equivalent to a gas velocity of approximately 34 cm/min. The steam flow rate in steam-air mixtures was four times that of air.

[b] Burnup level, 23.6 atom % ^{235}U. Specimens were in the form of 5/16-inch diameter disks punched from MTR-type fuel plates, re-irradiated to build up a suitable inventory of short-lived fission products.

[c] Air flow rate in these experiments was increased to 3000 cc/min or about 430 cm/min, measured at room temperature.

increase in tellurium and cesium release but the release of iodine and rare gases was so high with a short heating period that no time effect could be observed for these elements. The release values obtained for ruthenium in air were so low that no trend with either time or temperature could be established from the data obtained. The data in Table 4.9 also show that a drastic increase in air flow rate had little, if any, effect on fission-product release.

Data[43] on the effect of burnup on fission-product release from U-Al alloys at different temperatures are shown in Table 4.10. There is a noticeable burnup effect in the release of all the fission-product elements examined in these experiments but the largest and most important effect is in the release of the most volatile species, iodine and the rare gases. These data provide an explanation for the low results previously reported[8] for fuel specimens irradiated only to trace level. The data show little increase in release with increasing burnup above 3.2% except for cesium release at the highest burnup level. This fact seems to indicate that the burnup effect, whatever its explanation may be, is saturated at a comparatively low burnup level.

4.4 Release of Fission Products from Zirconium-Uranium Alloys

Data obtained[8] on melting zirconium-uranium alloy fuel specimens irradiated to a significant burnup level (15 atm % ^{235}U) in air and steam atmospheres are shown in Table 4.11. The apparatus used in melting experiments in air is shown in Fig. 3.1 while that used with steam atmospheres is shown in Fig. 3.2. These data confirmed the indication in previously published data[51] that the rare gases are quantitatively released but the iodine and cesium results are much higher on the average than those obtained with low-burnup fuel. The

Table 4.10. Effect of Temperature and Burnup on Fission Product Release from Irradiated Aluminum-Uranium Alloys[a]

Burnup Level (%)	Release (%)				
	Iodine	Tellurium	Cesium	Ruthenium	Rare Gas
At 750°C					
Trace	3.7	~0.01	~1.2	-	4.3
3.2	52.8	0.14	-	0.0005	98.2
9.0	54.2	0.05	1.3	0.004	-
23.6	56	~0.3	~3.6	0.07	~98
At 800°C					
Trace	16.7	0.02	1.6	~0.002	37.2
3.2	-	~0.3	1.1	0.01	99.4
9.0	71.9	0.04	1.7	~0.01	99.4
23.6	78.6	~0.5	3.8	0.08	99.4
At 900°C					
Trace	28.8	0.03	2.6	~0.004	54.0
3.2	97.4	6.1	2.7	0.002	>99.5
9.0	95.0	1.9	7.5	-	>99.5
23.6	92	2.0	6.2	0.1	>99.9
At 1000°C					
Trace	41.2	0.14	-	-	-
3.2	98.2	5.3	3.2	0.04	~100
9.0	97.2	6.3	3.5	0.2	~100
23.6	97.3	9.7	8.8	0.25	99.8
At 1100°C					
Trace	(34.3)	1.3	~6	-	71.8
3.2	99.5	31.7	9.5	0.03	~100
9.0	93.5	37.1	19.6	0.25	~100
23.6	98.4	~50	12.4	0.6	100

[a] Specimens were heated for 2 min at maximum temperature in air flowing at 250 cc/min.

Table 4.11. Fission-Product Volatilization from Melted Encapsulated Zircaloy Punched Disks, 15% Burnup

Run No.	Atmosphere During Melting	Heating Time[a] (sec)	Max Temp.[b] (°C)	Percent of Total Activity Released						
				Rare Gases	Gross γ	I_2	Cs	Sr	Ba	Ce
10-10	Air	12.5	1705	100	2.0	28	10			
10-15	Air	16	-	100	2.2	32	11			
10-16[c]	Air	12	1750	100	6.4	-	12			
11-25	Air	30	1705	100	2.5	14	7.3	0.9	0.1	0.005
11-26	Air	30	1800	100	4.1	30	13	0.8	0.3	0.004
	Av	20	1740	100	3.4	26	10.6	0.85	0.2	0.004
10-20	Steam	33		100	5.3	13	8.9	2.4	0.3	0.05
10-22	Steam	32	1775	100	5.5	52	23	3.1	1.2	0.01
11-11	Steam	18		100	7.8	66	22	4.8		
11-12	Steam	31	1750	100	7.2	56	24	1.3		
11-14	Steam	35	1750	100	4.8	57	13	0.2		
11-21	Steam	30.5	1750	100	8.0	45	19	0.2	0.05	
11-24	Steam	31.5	1730	100	5.7	36	20	0.8	0.2	
	Av	30	1750	100	6.3	47	19	1.8	0.4	0.03

[a] Sample usually melted in approximately 12 sec. [b] Optical pyrometer temperature.

[c] Punched disk not re-irradiated.

release of less volatile elements such as strontium, barium, and cerium was unaffected by burnup, which is in line with uranium-aluminum alloy experience.

It would be highly desirable, in view of continuing use of zirconium-uranium alloys in reactors, to supplement the early release experiments and to examine the distribution of the volatilized fission products in some detail.

5.0 INVESTIGATIONS WITH OXIDES DISPERSED IN A METALLIC MATRIX

5.1 Uranium Dioxide in Aluminum

There is very little data available on the release of fission products from UO_2 dispersed in aluminum. Creek et al.[8] reported results obtained with three tracer-level irradiated Geneva reactor fuel samples. An average of 5.6% of the rare gases and 0.003% of the iodine was released on melting the samples in air. The melting point of aluminum (659°C) is low enough so that diffusion of fission products from UO_2, even in the form of small particles, would not be expected to be great enough to be of significance in reactor accidents.

5.2 Uranium Dioxide in Stainless Steel

Studies of fission-product release from UO_2 dispersed in stainless steel[8] (Army Power Package Reactor or SM-1) fuel coupons have been made with material irradiated at trace level (Table 5.1) and with 20 to 30% burnup fuel (Tables 5.2, 5.3, and 5.4)using apparatus of the type shown in Figs. 3.1 and 3.2. The data in Tables 5.3 and 5.4 show a definite correlation between fission-product release and preheat time (time required to heat the fuel specimen from room temperature

Table 5.1. Fission-Product Volatilization from APPR Clad Coupons[a] Melted in Air or Steam

Run No.	Atmosphere	Preheat Time To Melt (sec)	Percent Total Activity Released				
			Rare Gases	Gross γ	Iodine	Cs	Sr
1	Air	125	45	5.4	49		
2	Air	55	59	2.2	25		
3	Air	42	38				
4	Air	94	48	4	15		
5	Air	137	40	1.5	34		
6	Air	90	54	8	31	11	0.001
7	Air	144	61	6	41	13	0.001
8	Air	75	44				
9	Air	151	50	4.5	38		0.0001
10	Air	75	46	4.4	35	3.4	0.1
		Av	48	4.5	34	9.1	0.03
11	Steam		39	2.3	11.2	0.3	0.4

[a] Coupons irradiated in graphite reactor for one week.

Table 5.2. Fission-Product Volatilization by Melting APPR Punched Disks, 25% Burnup

Run No.	Atmosphere During Melting	Maximum Temp. (°C)	Time to Melt (sec)	Percent of Total Activity Released				
				Gross γ	Iodine	Cs	Sr	Rare Gases
1[a]	Air	1575	13	0.4	-	12	-	-
2[a]	Air	1650	15	0.2	-	5.8	-	-
3	Air	1575	47	17	-	68	-	-
4	Steam	-	45	17	-	75	0.06	-
5	Helium	1650	37	26	-	99.9	6.8	-

[a] Decayed through long cooling period, not re-irradiated.

Table 5.3. Variation in the Amounts of Iodine and Rare Gases Released from APPR Disks[a] with Different Preheat Times

Sample No.	Preheat Time (sec)	Pyrometer Reading at m.p. (°C)	Percent of Total Activity Released	
			Rare Gases	Iodine
1	17	1300	16	3.7
2	20	1565	31.3	5.7
3	31	1500	19.4	15.2
4	59	1618	44.6	15.3
5	61	1550	41.0	15.9
6	72	1521	50.7	17.6
	Av 43.3		34.8	12.2

---

[a] 20% burnup.

Table 5.4. Variation in the Amount of Cesium Released from APPR Disks[a] with Different Preheat Times

Sample No.	Thickness of Heater (in.)	Preheat Time (sec)	Percent 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

---

[a] 30% burnup.

to the melting point of the cladding and matrix material). The rare gas and iodine data display considerable scatter but the correlation is somewhat better for gross gamma and cesium data (Fig. 5.1).

The higher melting point of the stainless steel results in higher release of volatile fission products from the melted fuel, as compared to that from UO_2 dispersed in aluminum. Little difference was noted in release values in air and steam but the release of cesium and strontium was higher in helium than in oxidizing gases, reflecting the higher volatility of the element as compared to the oxide. At the maximum temperatures attained in these experiments (1575 to 1650°C), Cs_2O would be largely dissociated but the fuel specimens were at this temperature for only a fraction of the total heating time.

5.3 Uranium Dioxide in Nichrome

Fuels consisting of uranium dioxide dispersed in Nichrome V were considered for use in the direct-cycle-reactor system at the Aircraft Nuclear Propulsion Project at one time and a few experiments were performed[8] to determine the extent of fission product release from fuel specimens of this type. The data given in Table 5.5 were obtained with trace-level irradiated fuel specimens and the values are not significantly different from those obtained with trace-irradiated stainless steel-uranium oxide dispersions under comparable conditions. Iodine and cesium release values are lower than those obtained with high-burnup fuel materials. A more detailed examination of release from this type of fuel may be required if use of Nichrome-UO_2 dispersions in power reactors is contemplated.

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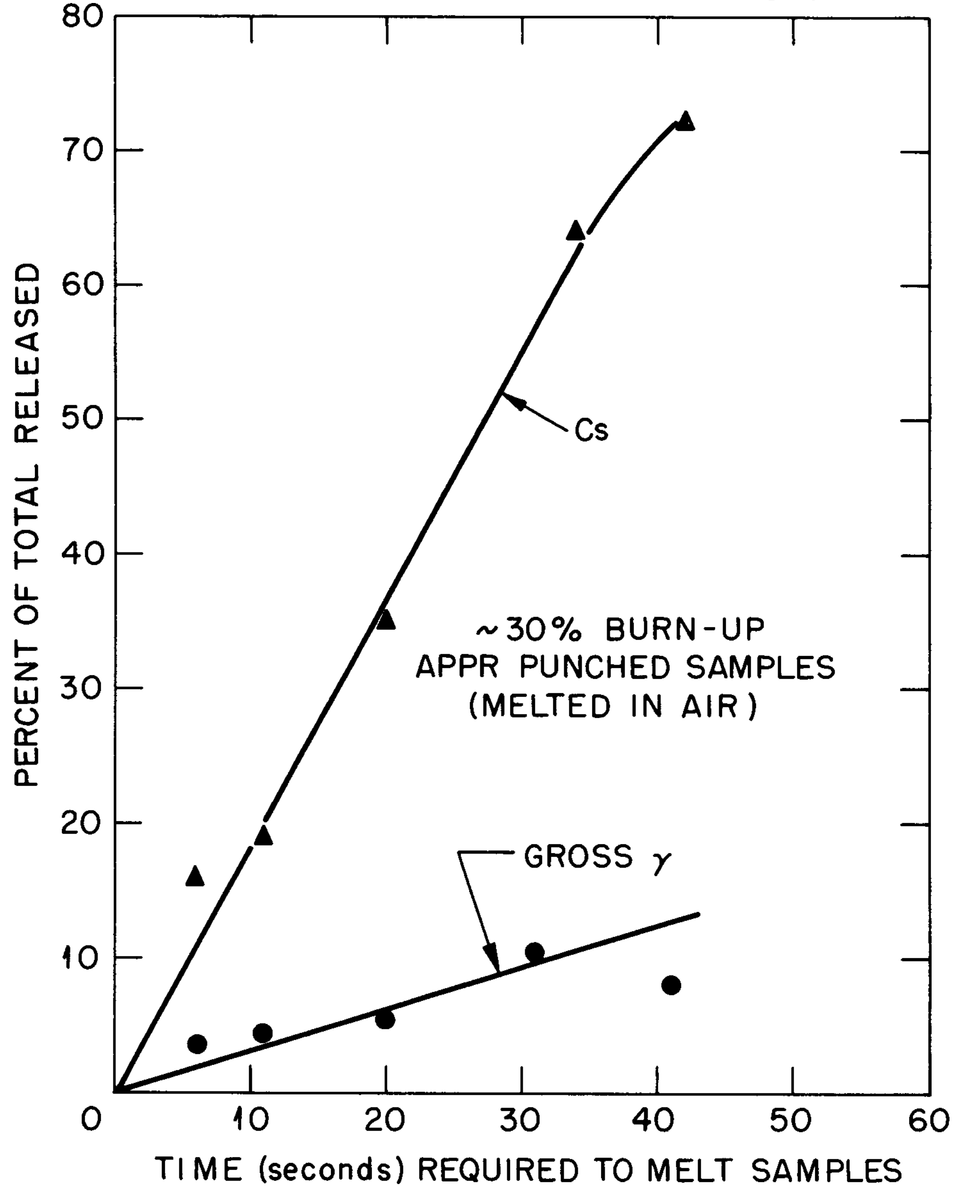

Fig. 5.1. Correlation of Per Cent Release of Cesium From SS-UO_2 Dispersion Fuel with Total Heating Time.

Table 5.5 Release of Fission Products From Nichrome V-Uranium Dioxide Dispersions Heated in Air

Heating Time	Percent of Total Fission Product Activity Released			
	Rare Gases	Iodine	Cesium	Strontium
30 sec	9.7	4.2	0.7	
32 sec	10.2	4.4	0.3	0.0001
4 hr	77	99.1	1.7	0.01

6.0 INVESTIGATIONS WITH OXIDE FUEL MATERIALS

6.1 Uranium Dioxide, UO_2

Fission products may be released from UO_2 fuels during an accident by diffusion, oxidation, and melting in the approximate order of increasing extent of fission-product release (see Section 2). Each release mechanism is affected by many parameters and obtaining experimental data on all parameters under all conceivable accident conditions would obviously require a large amount of effort. Work in this field has been directed toward determining the relative importance of the parameters and toward evaluating their effect on fission-product release under most probable accident conditions. The various release mechanisms will be considered in the order indicated above.

6.1.1 Release of Fission Products by Diffusion

Release of fission products from UO_2 by diffusion has been studied by more investigators than of the other two mechanisms combined but it is still not thoroughly understood.

Studies [52,56] made by heating trace-irradiated PWR-type UO_2 pellets in a flowing stream of purified helium for 5.5 hr gave the data in Table 6.1. Apparatus shown in Fig. 3.7

Table 6.1. Diffusion of Fission Products from UO_2[a] into Purified Helium[b]

Temp. (°C)	Rare Gases	Gross γ	Percent Release							
			I	Te	Cs	Ru	Sr	Ba	Zr	U
1515	1.3	0.9	5.8	2.9	1.4	0.9	0.1			
1610	2.7	2.1	6.5	12	1.7	1.5	0.1			
1710	2.6	6.3	9.6	20	2.7	3.8	0.4	1.3		
1800	3.7	5.2	12	21	3.2	6.9	1.0			
1900	9.7	12	16	48	8.6	8.5	2.3			
1980	12	12	42	76	15	13	4.2	8.7	1.8	
2105	25	23	40	81	24	22	13	21	0.5	0.5
2150	59	18	74	95	53	49	28	40	12	0.4
2200	65	33	75	96	70	50	36	59	18	
2260	87	38	84	96	65	90	55	75	35	1.3

[a] Trace-level irradiated PWR-UO_2 samples heated 5.5 hours in tantalum crucibles by RF induction.

[b] Helium purified by contact with hot zirconium sponge; flow rate 50 cc/min.

was used in these investigations. The smaller fraction of the sample volatilized in these experiments, as compared to the British results[57] obtained at comparable times and temperatures, is possibly due to the difference in sample size (0.05 vs 7 grams). It is clear from these data that escape rates are high enough at temperatures of 1700°C and above to permit release of significant quantities of fission products over a period of hours. At 2100°C and above, even such low-volatility elements as barium and zirconium volatilize to a significant extent in 5.5 hours.

A plot of diffusion constant vs 1/°K for the diffusion of rare gas (Xe) from trace-irradiated PWR-type UO_2 is shown in Fig. 6.1. The inflection point at about 1800°C in this plot undoubtedly is due to grain growth and since this phenomenon is a function of both time and temperature, the slope probably does not change sharply.

The release of various fission-products during 5.5 hours heating of trace-level UO_2 is compared in Fig. 6.2. This shows that both iodine and tellurium diffuse at a faster rate than the rare gases. Similar plots for UO_2 irradiated to higher levels are given in Fig. 6.3.

The effect of burnup on release of fission products by diffusion at four temperatures is shown by the data in Table 6.2. Data from Table 6.1 are included for comparison although the samples were not identical even for PWR UO_2. None of the high burnup UO_2 materials received to date have included intact pellets such as those employed to obtain the data in Table 6.1. Fragments of variable size were, of necessity, employed to study the burnup effect. These fragments ranged in weight roughly from 0.1 to 0.2 grams and sample weights varied from about 1 to 2 grams. There is no clear-cut burnup effect evident at lower temperatures (1400 or 1610) or at low burnups (1000 Mwd/ton) but otherwise it is quite plain that diffusion of fission products increases with increasing burnup, except possibly for the low-volatility elements.

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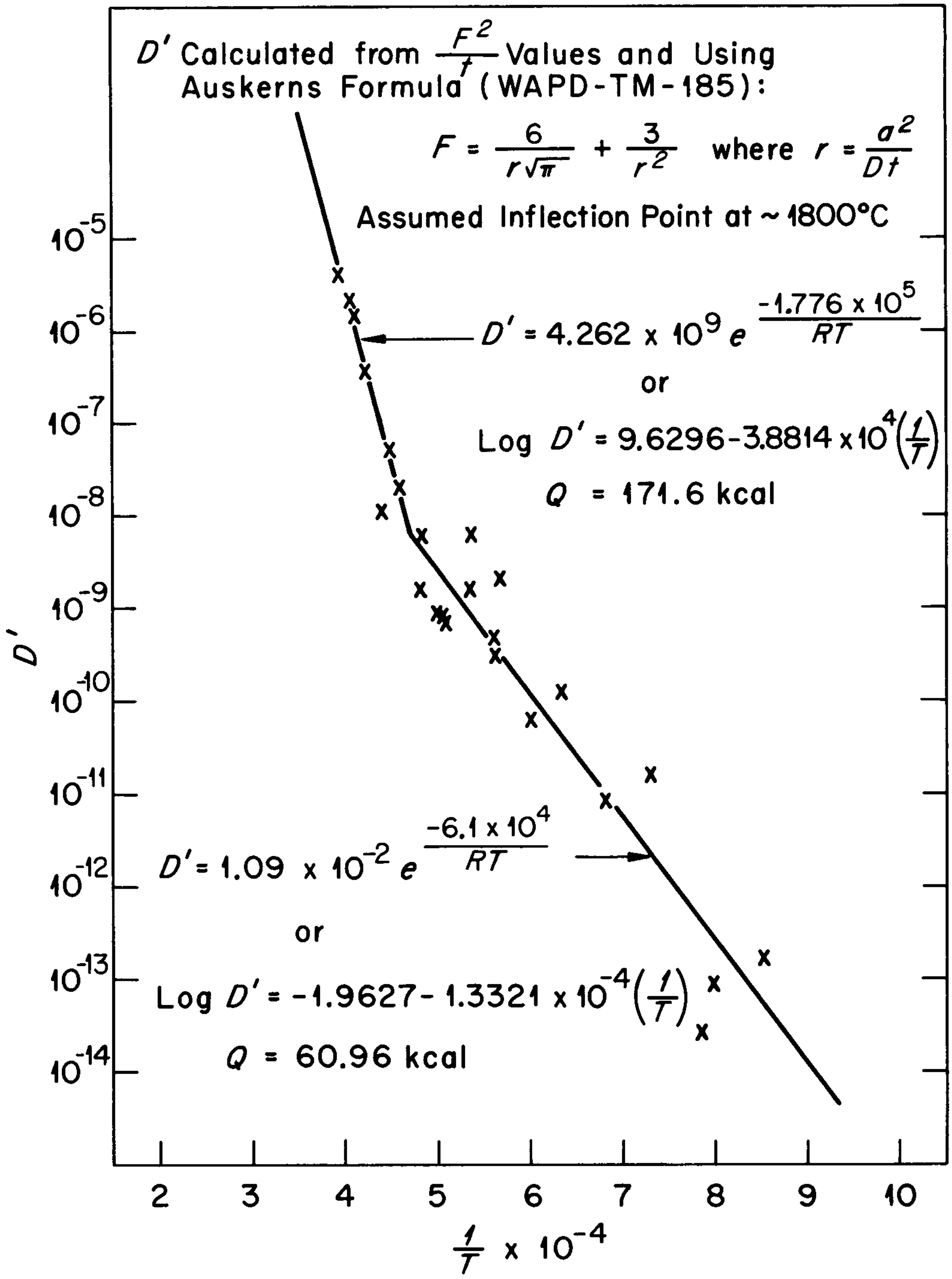

Fig. 6.1. Rare Gas Diffusion from PWR UO_2 in Helium.

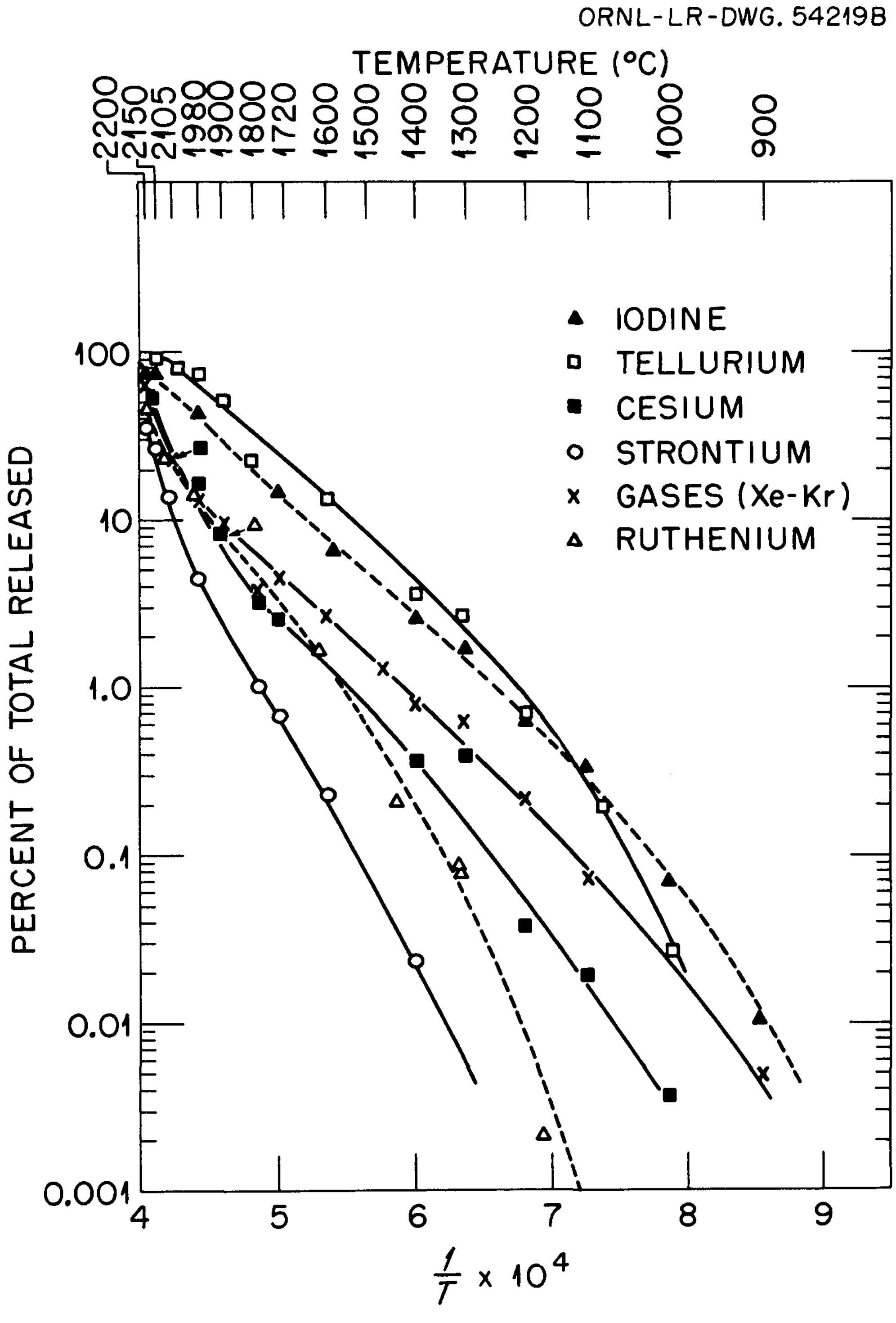

Fig. 6.2. Release of Fission Products by Diffusion from Trace-Level Irradiated UO_2 Heated in Pure Helium for Five Hours.

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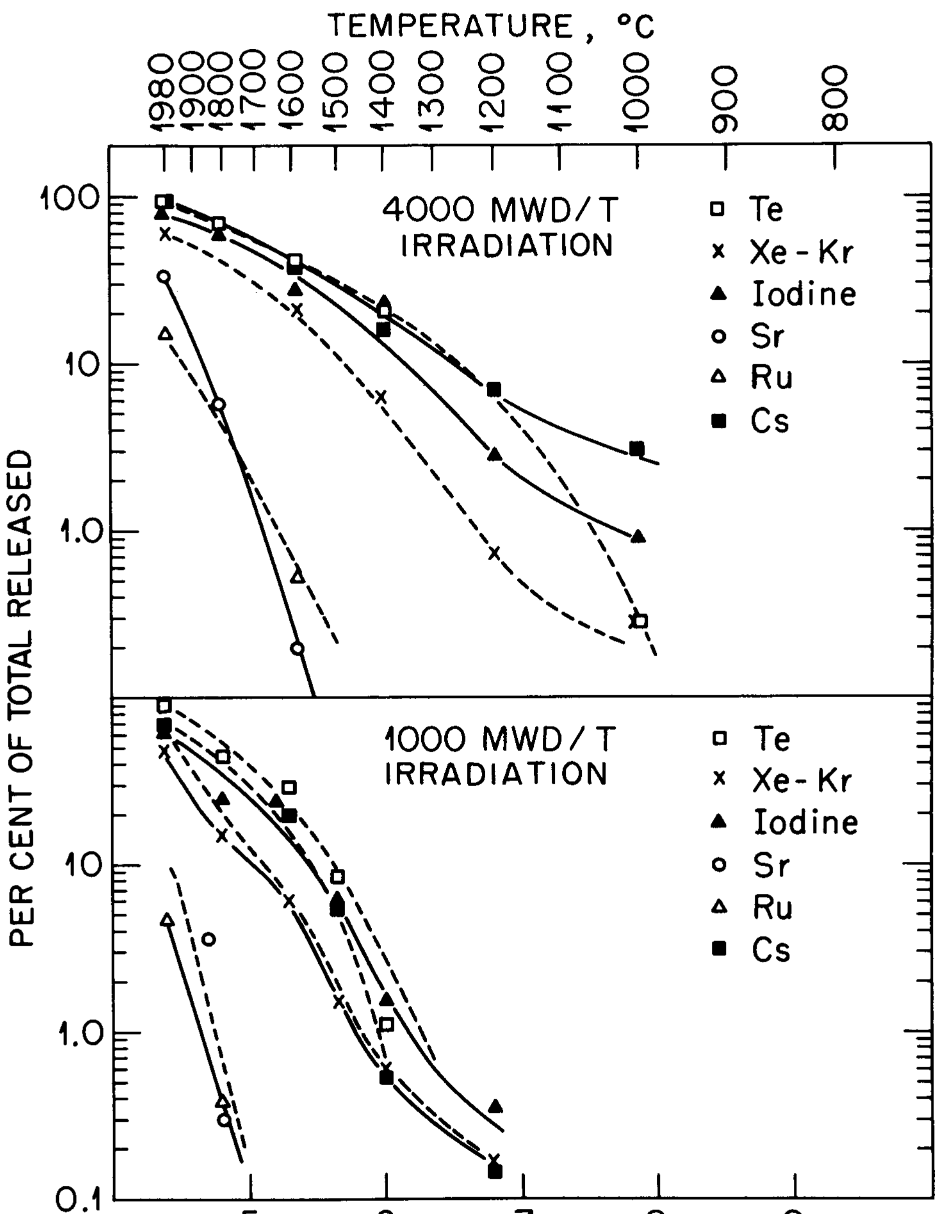

Fig. 6.3. Release of Fission Products by Diffusion from Highly-Irradiated PWR-Type UO_2 Heated 5.5 Hours in Purified Flowing Helium.

Table 6.2. Effect of Burnup Level and Temperature on Diffusion of Fission Products from UO_2 Heated 5.5 Hours in Pure Helium

Temperature (°C)	Irradiation Level (Mwd/ton)	Percent of Individual Fission Products Released[a]						
		Xe-Kr	I	Te	Cs	Ru	Sr	Ba
1400	~1[b]	0.8	4.0	3.9	0.02	0.02	0.001	
	1005[c]	0.8	0.9	0.8	2.6	0.001	0.1	
	1000[b]	0.5	1.6	1.2	0.5	0.001	0.06	1.8
	4000[b]	6.1	23	16	21	0.006	0.08	0.5
1610	~1[b]	2.7	6.5	12	1.7	1.5	0.1	
	1005[c]	2.6	3.7	12	12	0.1	2.0	17
	1000[b]	6.0	5.5	27	20	0.3	0.2	12
	4000[b]	14	25	48	43	0.2	0.5	15
1780	~1[b]	3.7	12	21	3.2	6.9	1.0	
	1005[c]	12	24	67	27	0.4	9.0	39
	1000[b]	14	26	35	22	0.4	3.7	21
	4000[b]	42	59	60	40	5.7	5.8	18
1980	~1[b]	12	41	75	15	13	4.2	8.7
	1005[c]	29	53	74	84	6.0	15	57
	1000[b]	49	63	90	70	4.8	~10	51
	4000[b]	71	81	81	98	15	33	60

[a] Includes that portion adsorbed on crucible and reflector parts.

[b] PWR-type UO_2 (93 to 94% of theoretical density) - only the 1 Mwd/T pellets were full size (7 gm). The high-burnup samples were 0.1-0.2 gm fragments with a total weight of 1 to 2 gm.

[c] EGCR-type UO_2 (97% of theoretical density). Samples were similar to PWR samples in total weight and fragment size.

Data on diffusion rates at temperatures between 2260°C and the melting point of UO_2 (2860°) are very scarce. Release results obtained in a transient reactor experiment[58,59] where the specimen reached a temperature above 2600°C for a very short time (seconds) but did not melt, indicated that release rates of the more volatile elements are very high in this temperature range.

One type of release from UO_2 that has received comparatively little attention in comparison to other release mechamisms is the escape of fission gases resulting from rapid cooling.

Parker and co-workers[55] compared the gas release behavior of trace-irradiated and highly-irradiated UO_2 specimens on heating to 1600°C and cooling. The results are shown in Fig. 6.4. This temperature is probably too low for information of appreciable amounts of substoichiometric oxide, which probably accounts for failure to observe significant release on cooling the trace-irradiated material, but the reason for the large cooling release in the highly irradiated specimen is not clear. A similar release is shown in Fig. 4.11. It has not been established whether other fission products show a cooling burst effect but the possible contribution of this effect to the overall hazard of loss-of-coolant accidents needs consideration if reactor accident conditions permit rapid cooling of overheated fuel.

6.1.2 Oxidation of Uranium Dioxide

A comparison of initial oxidation rates of specimens differing only slightly in density is shown in Fig. 6.5. Oxidation rates were determined in the same apparatus (Fig. 3.4) used for oxidation of metallic uranium. Surface areas plotted are geometrical areas. Agreement with data obtained by Peakall and Antill[60] seems quite good. It is clear from this figure that oxidation rates are strongly dependent on density and on surface area, which probably explains some of

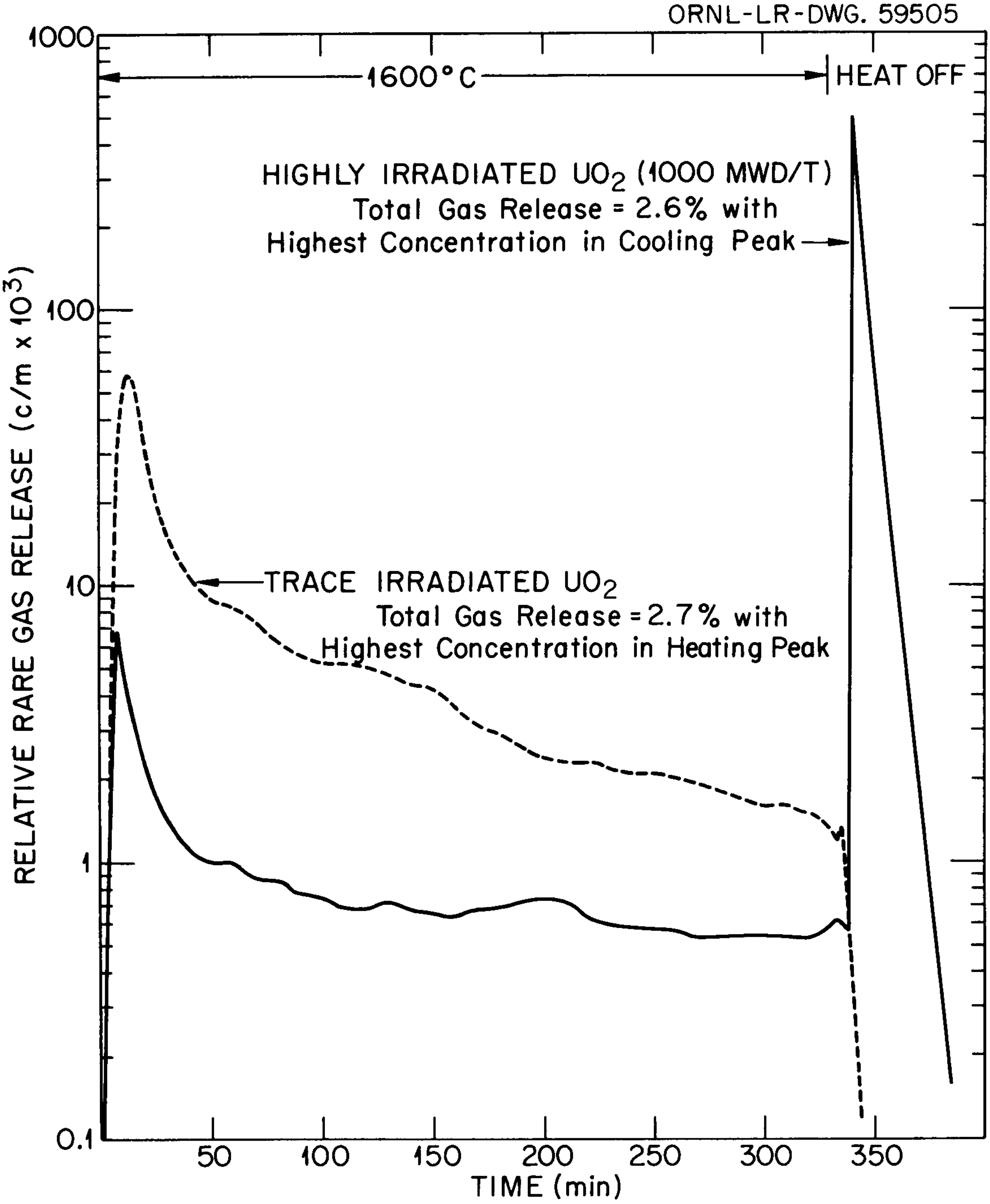

Fig. 6.4. Comparison of Rate of Release of Rare Gas from Trace-Irradiated and High-Burnup UO_2

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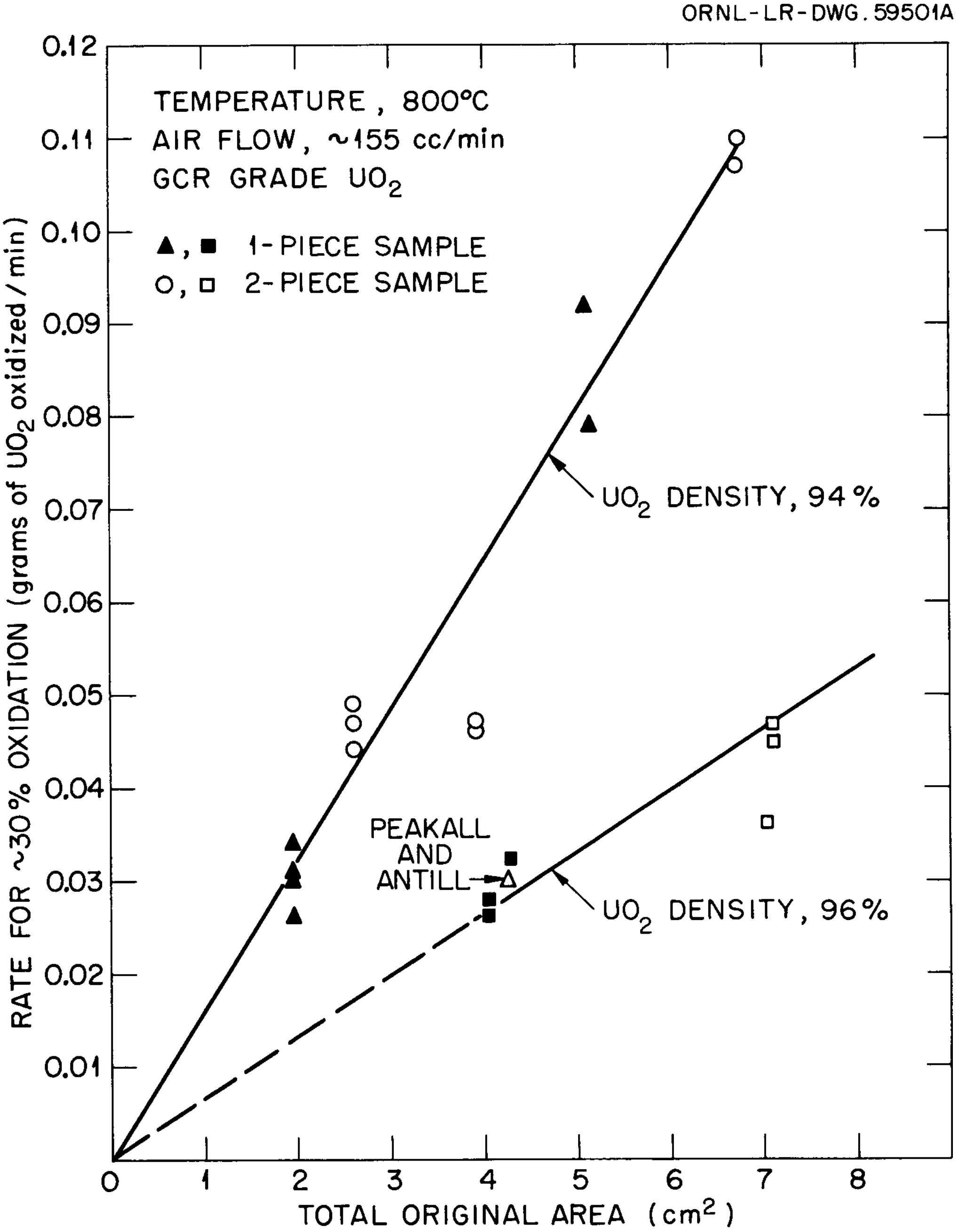


Fig. 6.5. Effect of Density on Rate of Oxidation of UO_2.

the confusion that has arisen in regard to oxidation rates.

Data on the effect of varying furnace temperature on the rate of oxidation of UO_2 in air are shown in Figs. 6.6, 6.7, and 6.8.

Oxidation of UO_2 by steam apparently does not occur at significant rates. The sintering effect of steam on powdered UO_2 at moderately high temperatures (800 to 1000°C) was mentioned earlier in this report (Section 4.1.3). Data on rates of oxidation of UO_2 in steam-air mixtures seem to be lacking at present.

The complexity of the oxidation of UO_2 in air makes it desirable to have oxidation rates with full-size, highly-irradiated fuel elements and to obtain fission-product release data concurrently with the oxidation studies. Such investigations can obviously be performed only in well designed and well shielded hot-cell facilities.

6.1.3 Fission-Product Release Resulting from Oxidation of UO_2

Fewer studies of fission-product release accompanying the oxidation of UO_2 have been made than of diffusion release. Parker and co-workers[53] first studied release from trace-irradiated PWR-type UO_2 pellets of 94% of theoretical density and later[54] from the same type of fuel irradiated to different levels of burnup to a maximum of 7000 Mwd/ton. Release values determined with specimens irradiated at trace level (~1 Mwd/T) are plotted in Fig. 6.9, while data obtained at the highest burnup level are given in Table 6.3. It is clear from these data that fission product release is not a simple function of temperature. This complex behavior might be expected from the oxidation data shown in Figs. 6.6, 6.7, and 6.8.

The effect of varying the heating time at different temperatures is shown in Table 6.4. Specimens employed to obtain these data were PWR-type material with a density of

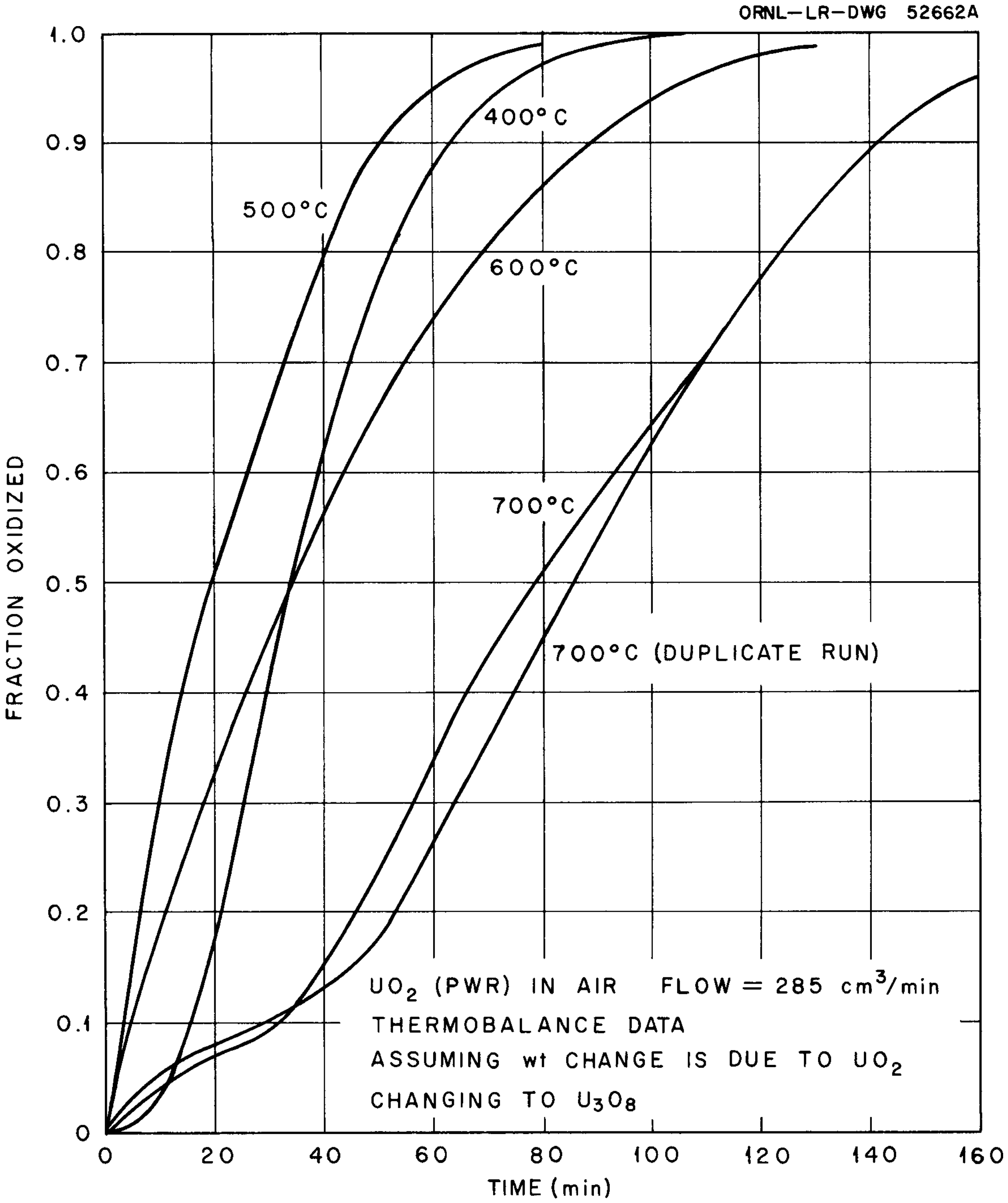


Fig. 6.6. Rate of Oxidation of UO_2 in Air at 400 to 700°C.

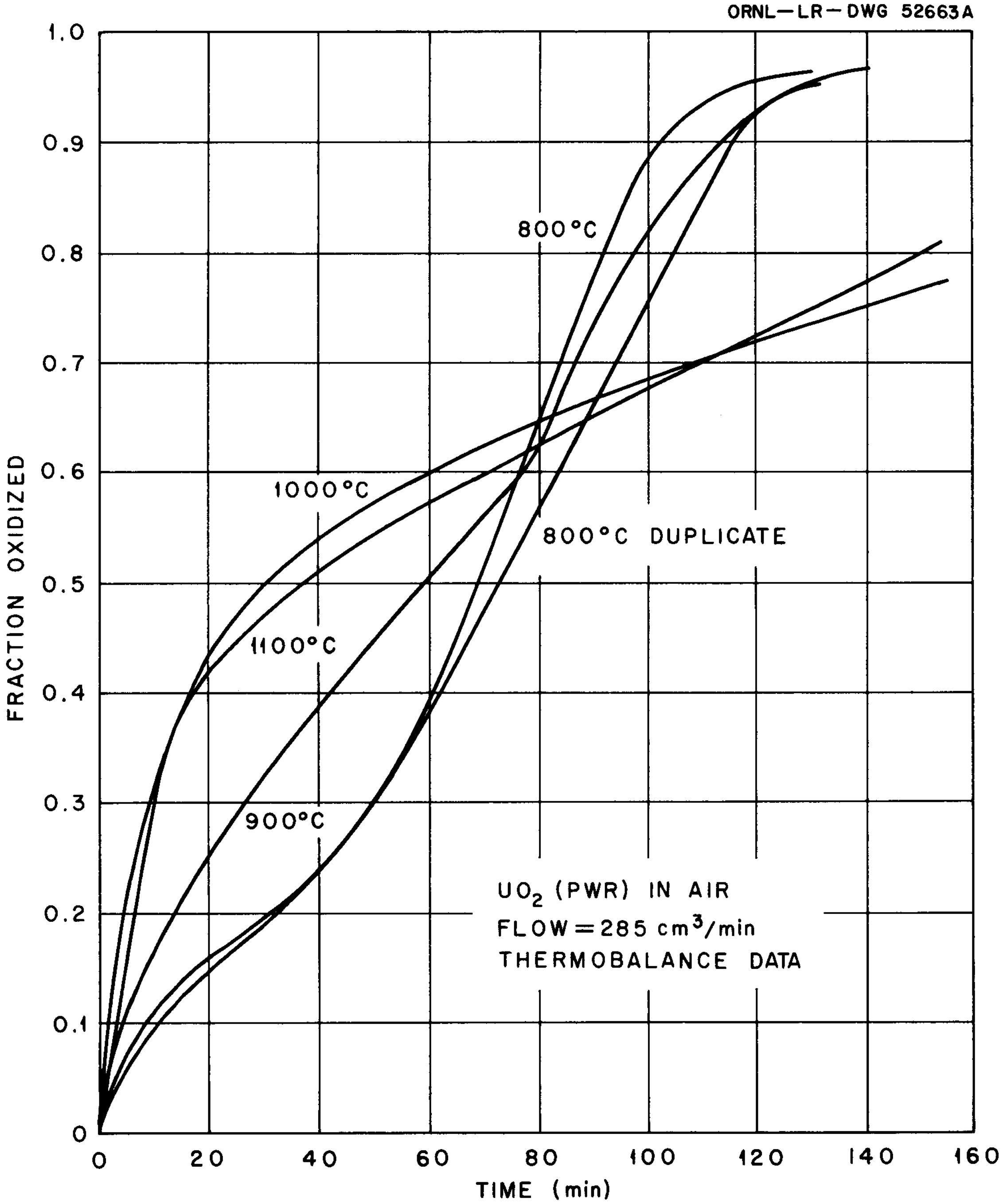


Fig. 6.7. Rate of Oxidation of UO_2 in Air at 800 to 1100°C.

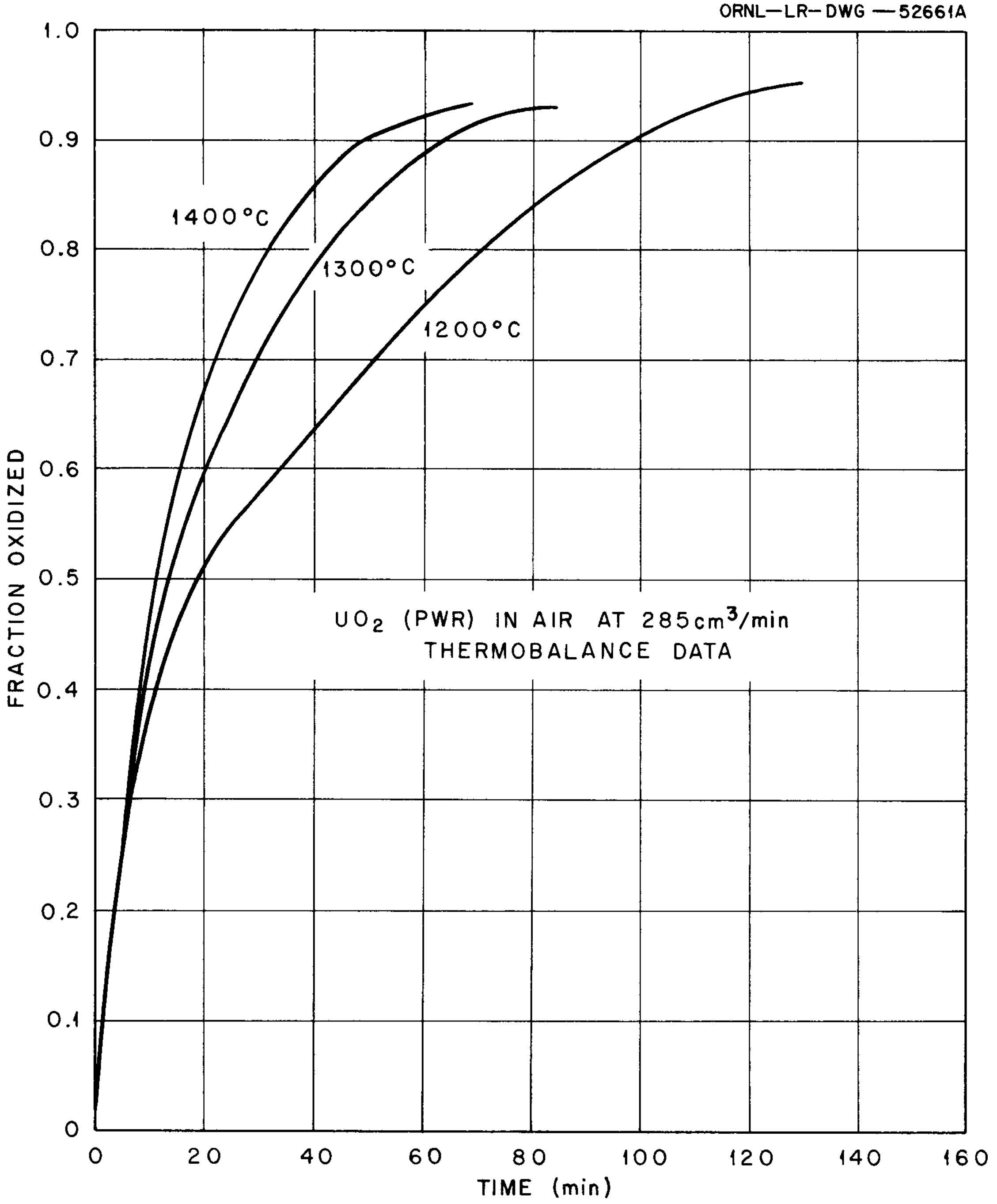


Fig. 6.8. Rate of Oxidation of UO_2 in Air at 1200 to 1400°C.

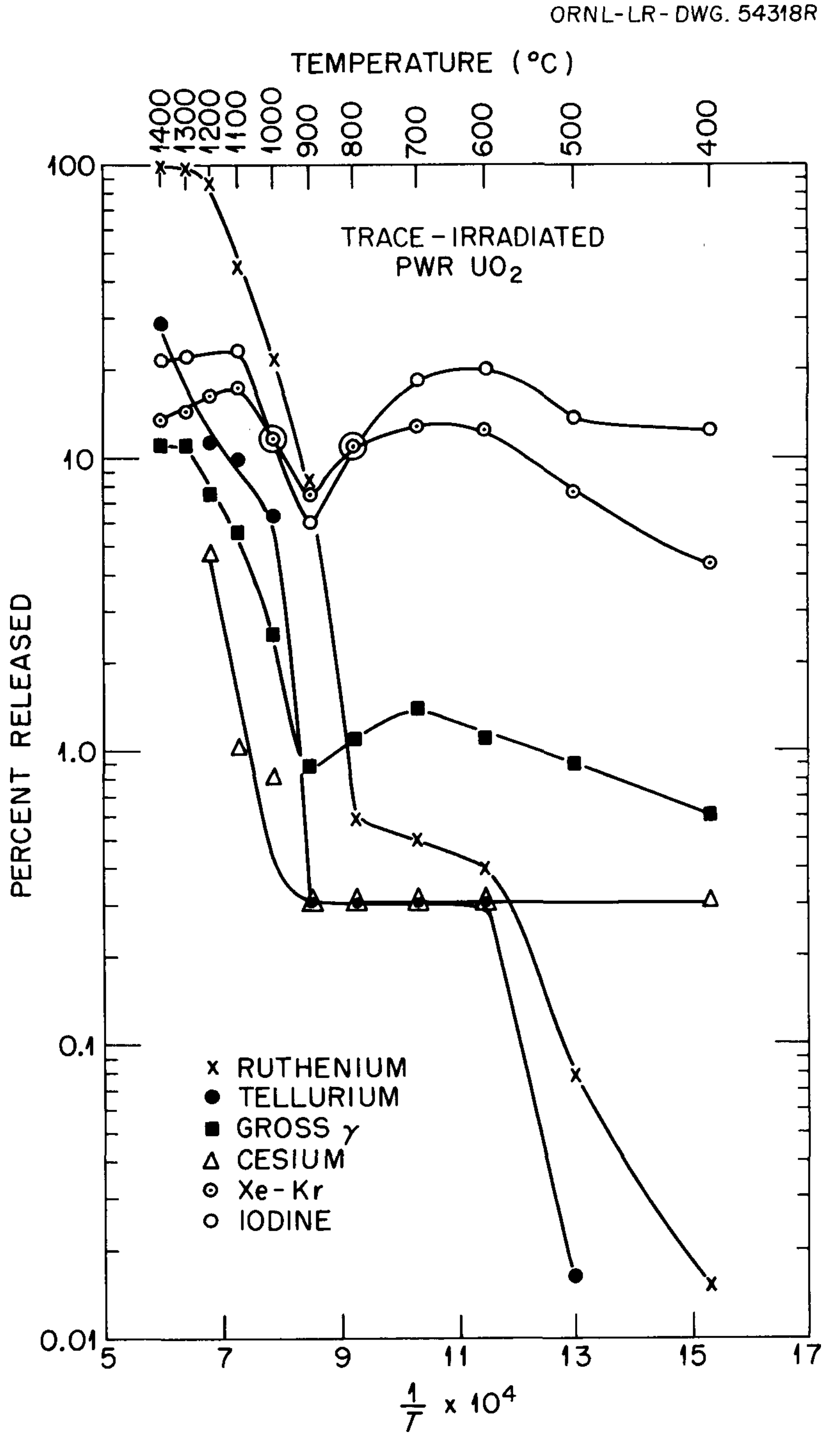


Fig. 6.9. Fission Product Release by the Oxidation of UO_2 to U_3O_8 in Air, Showing Discontinuity Between 600° and 900°C.

Table 6.3. Fission-Product Release from PWR-Type UO_2[a] Irradiated to 7000 Mwd/T and Heated in Air[b] for 90 min.

Temp. (°C)	Percent of Individual Fission Products Released						
	Rare Gases	I	Te	Cs	Ru	Sr	Ba
500	3.1	4.1	<0.5	0.0006	0.1	<0.0007	<0.0004
600	4.2	3.1	<0.1	<0.002	0.7	<0.0004	<0.009
700	6.1	15	<0.08	<0.005	0.1	<0.0005	<0.007
800	9.4	9.0	<0.3	0.002	9.8	<0.0005	0.03
850	15	34	1.4	0.02	35	<0.005	<0.08
900	34	29	80	<0.01	78	<0.03	<0.8
1000	86	78	37	<0.03	93	<0.04	<0.3

[a] Samples 0.5 to 0.9g of 96% density material in porous alundum cups, preheated for 13 to 16 min. in helium.

[b] Air flow, 100 cc/min.

Table 6.4. Fission-Product Release from PWR-Type UO_2[a]
Irradiated to 4000 Mwd/T and Heated in Air[b]

Temp. (°C)	Time at Temp. (min.)		Percent of Individual Fission Products Released							
	He	Air	Rare Gases	I	Te	Cs	Ru	Sr	Ba	U
500	16	23	1.5	3.6	<0.007	<0.0004	<0.005	<0.0004		
	18	90	2.9	3.2	<0.01	<0.0007	<0.01	<0.0004	<0.0008	
600	14	18	4.4	10	<0.006	0.002	0.08	<0.001		
	15	90	4.5	8.0	8.4	<0.001	1.8	<0.001	<0.004	
700	14	12	9.3	9.6	0.01	0.001	1.7	<0.0002	<0.0004	
	13.5	15	7.0	10	0.004	<0.001	0.4	<0.0003	<0.0006	
	14	90	6.8	6.5	<0.05	<0.0005	2.3	<0.0004	<0.002	
800	13	15	14	7.1	0.007	0.015	1.0	<0.0004	<0.0007	
	14	90	14	16	<0.06	<0.01	12	<0.0004	<0.001	
900	14	19	21	49	0.4	0.009	17	<0.001	0.01	
	15	90	22	47	6.0	0.015	53	<0.0008	<0.004	
1000	16	15	40	84	12	0.09	72	<0.0003	<0.02	
	13.5	90	44	75	32	0.37	92	0.1	0.08	0.06
1100	14	14	66	79	16	<0.02	91	<0.05	<0.003	
	14	90	73	84	39	0.2	99	0.006	0.01	<0.003
1200	14	16.5	71	82	37	0.8	99	<0.01	<0.001	
	13	90	80	95	66	6.4	99.6	0.007	0.7	<0.003

[a] Sample approximately 1 g of intermediate density (93 to 94%) material in porous alundum cups.

[b] Air flow, 100 cc/min.

93 to 94% of theoretical, irradiated to a burnup of 4000 Mwd/T. Increasing exposure time in air in the range investigated seemed to have no significant effect on fission-product release below 800°C but at this temperature and above, increasing release of some isotopes with increasing exposure was observed as might be expected. Similar data obtained with lower burnup material are contained in Table 6.5.

The effect of burnup on oxidation release of the more volatile fission products is shown graphically in Fig. 6.10 for two temperatures. The largest effect in the case of iodine and ruthenium came in the first 1000 Mwd/T of burnup and this was also true of the rare gases at 1200°C. The release of tellurium appeared to increase more or less regularly with increasing burnup in the range tested. The release of cesium, even at 1200°, was too low to establish an unequivocal correlation but the results obtained indicate a slight increase in release with increasing burnup.

The outstanding feature of these results is the high release of iodine and ruthenium. Conditions for the release of the latter appear to be especially favorable indicating that this element is readily converted to a volatile oxide.

6.1.4 Fission-Product Release From Molten UO_2

Melting of irradiated UO_2 is the most drastic mechanism for releasing fission products but, on account of the very high melting point of UO_2, it is also the least likely to occur. The principal parameters investigated were burnup, sample size, time molten, atmosphere, melting method, and type of cladding. Most of the melting experiments were performed in helium because of reactivity of container or heater materials with oxygen but it seems probable that the atmosphere surrounding molten UO_2 has little effect on the extent of release. A few experiments were performed with CO_2, air, and steam-air mixtures. There can be little doubt, however, that the atmosphere will have a drastic effect on post-release

Table 6.5. Fission Product Release from UO_2 Oxidized in Air

Sample: Intermediate density PWR UO_2 (93-94%)
Irradiation: 1000 Mwd/ton
Air flow: 100 cc/min

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

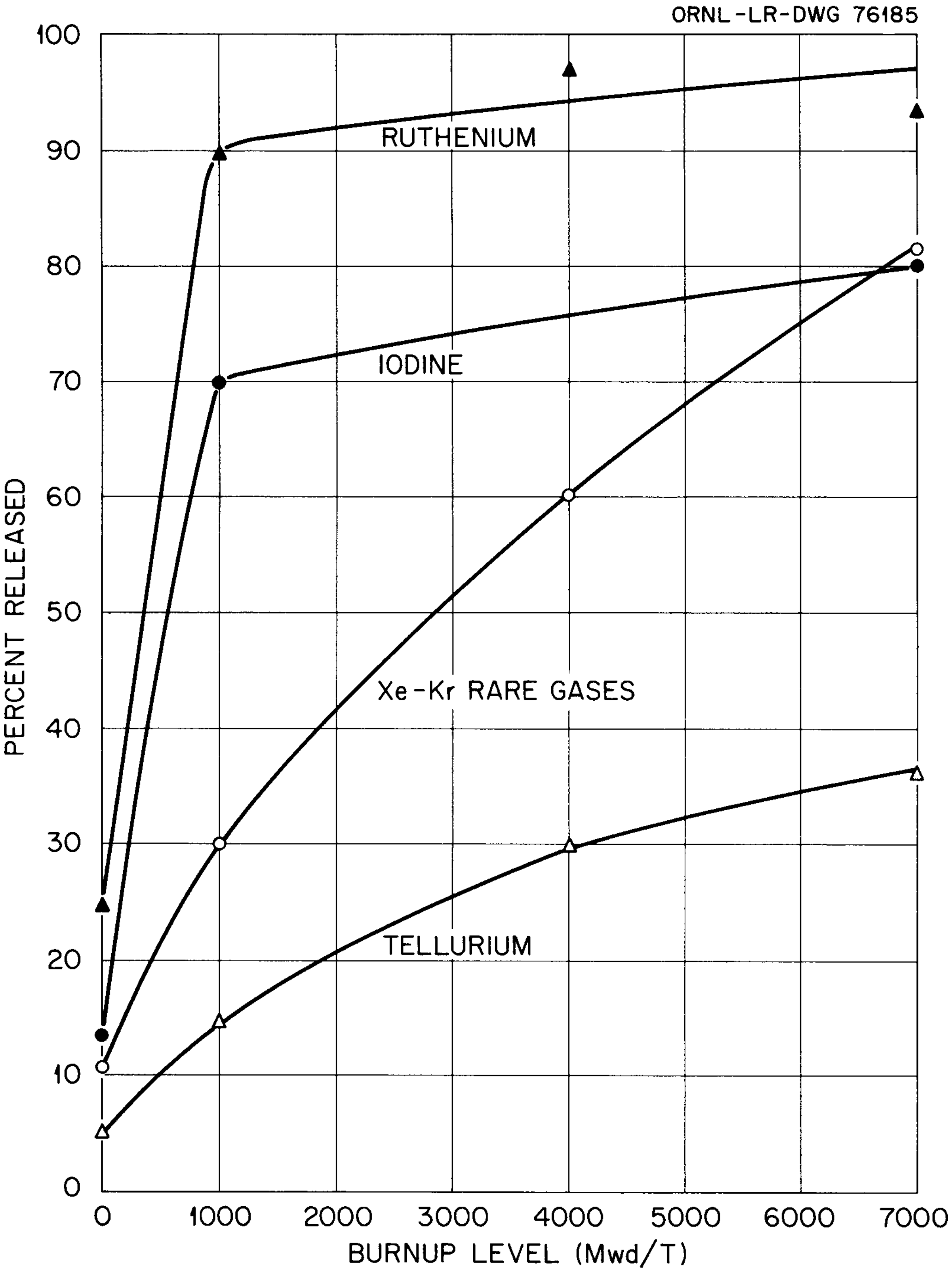


Fig. 6.10. Effect of Burnup on Fission-Product Release by Oxidation at 1000° and 1200°C.

behavior of the more reactive fission-product elements. During most of the period covered by this report the only reported experiments on measurement of fission-product release from molten UO_2 were performed at the Oak Ridge National Laboratory and a variety of methods have been employed in these investigations.

Results of the first melting experiments[53] with small trace-irradiated UO_2 specimens melted in an arc-image furnace (Fig. 3.5) along with its BeO support tube are given in Table 6.6. The sample support and the fission product collection train used in these experiments are shown in Fig. 3.6. The only parameters varied were the sample weight and the melting time. The data show that for these small specimens, release of all fission products sought, except for strontium, barium, and rare earths, was very high when complete melting of the specimens was achieved. Results of other experiments with different atmospheres and three levels of burnup are shown in Table 6.7. There appears to be a definite increase in release between trace-level irradiation and 2800 Mwd/T but there were no significant differences between the results at 2800 and 11,000 Mwd/T. There is no evidence in these data that the atmosphere surrounding the molten specimens affected the extent of release.

Data[56] obtained with larger samples of trace-irradiated UO_2 heated in tungsten crucibles (Fig. 3.7) are given in Table 6.8. The time that the UO_2 remained molten varied to some extent in these experiments and there seems to be a positive correlation between release and time molten for the more volatile elements. Comparable release results were, in general, obtained by the two melting techniques except for ruthenium. This element is known to be quite oxygen sensitive and it is quite possible that its higher release on melting in the arc-image furnace can be attributed to traces of oxygen in the helium supply and the absence of the good oxygen getter

Table 6.6. Fission Product Release from UO_2[a] Melted in Helium[b]

Run No.	Sample Weight (g)	Time at Temp. (sec)	Percent Release								
			Rare Gases	Gross γ	I	Te	Cs	Ru	Sr	Ba	TRE[d]
1	0.57	120	64	9.4	71	60	59	28	0.18		
2	0.34	120	91	13	70	72	25	60	0.07	0.8	0.2
3	0.56	120	93	6.0	84	86	34	32	0.16	0.9	1.1
4	0.56	180	56	7.7	67	63	24	75	0.11	1.3	0.7
5	0.58	180	63	14	46	54	12	36	0.11	2.6	0.5
6	0.37	120	69	10	51	43	7.1	20	0.26	0.5	0.3
7	0.18	120	99.4	30	84	86	90	72	0.20	2.0	0.7
8	0.25	90	99.6	31	95	96	93	76	3.9	7.3	3.8

[a] Trace-irradiated pellet melted simultaneously with BeO support tube in arc-image furnace.

[b] Helium flow rate, 100 cc/min.

Table 6.7. Effect of Irradiation and Atmosphere on Fission Product Release Resulting from the Melting of UO_2[a]

Atmosphere	Irradiation Level (Mwd/T)	Wt. of Sample (g)	Percent of Individual Fission Products Released								
			Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Rare Earths	UO_2 Vaporized
Helium	Tracer	0.22[b]	99.5	90	92	91	61	2.1	4.5	2.2	
(Impure)	2800	0.03	99.9	92	98	99	90	2.1	6.6	5.1	21
Air	Tracer	0.2[b]	98	95	79	38	68	0.2	0.5	0.5	
	2800	0.04	100	99.7	94	93	95	0.4	1.8	3.0	
CO_2	Tracer	0.2	81	77	71	61	45	0.3	1.1	0.9	14
	2800	0.02	99.9	99	99	90	74	0.5	2.5	2.8	
	11000	0.05	99.9	99.9	99	97	79	0.6	2.9	2.3	

[a] EGCR UO_2, with O/U ratio of 2.04 and density 95% of theoretical (average), melted in arc-image furnace.

[b] Average of two results; all others are averages of three results.

Table 6.8. Fission Product Release from UO_2[a] Melted in Helium[b] by the Tungsten-Crucible Method

Molten Time (min.)	Percent UO_2 Vaporized	Gross γ Release (%)	Percent of Individual Fission Product Released							
			Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Ce
1.0	0.10	23	93	77	90	63	0.45	0.33	4.8	0.05
1.5	0.16	15[c]	98	98	98	66	0.05	0.47	2.6	0.07
2.0	0.16	26	99	99	99	60	0.32	0.41	3.0	0.17
2.5	0.25	13[c]	99	95	99	72	0.33	0.53	2.4	0.13
1.5[d]	-	14[c]	99	88	92	80	0.20	0.26	2.6	0.40
2.5[d]	-	13[c]	99	93	96	89	0.70	0.50	3.6	0.10

[a] Sample: 29g PWR UO_2 irradiated at tracer level and preheated in helium for 4.5 to 5.0 min.

[b] Atmosphere: purified helium flowing at a rate of 700 cc/min.

[c] Decayed 4 to 7 days longer than previous sample.

[d] UO_2 sample had a slightly higher density than the first four samples.

(tungsten) that was available in the later experiments with larger samples.

Results of a third type of melting experiment[56,61] in which a tungsten rod resistor passed through cored UO_2 pellets served as the heating element (See Fig. 3.9) are shown in Table 6.9. Both clad and unclad elements were employed in these experiments, but they were limited to a helium atmosphere and complete melting of specimens could not be accomplished before the tungsten rods melted. Nevertheless, the results are quite useful because the high interior fuel temperature and cooler surface achieved with this heating method more nearly simulate nuclear heating than any other out-of-pile technique.

Release data obtained in single-pin experiments are compared in Table 6.10 with the results of two experiments with clusters of seven pins (Fig. 3.10). The center pin was irradiated to trace level (10^{14} fissions/gram of UO_2).

Table 6.10. Release From Center-Resistor-Heated Bare UO_2 Fuel Pins Corrected to 100% of Melting

No. of Fuel Pins	Release %							
	UO_2	I	Te	Cs	Ru	Sr	Ba	Ce
1	0.8	70	90	82	1.1	2.5	9.0	<1.0
7	3.1	60	45	37	6.3	1.1	1.9	0.1

The results in Table 6.9 obtained with unclad specimens[54] after adjusting for the fraction of the fuel melted, are not drastically different from the results of the other melting methods, except that release of strontium and barium was rather high and low ruthenium release was observed, indicating absence of free oxygen. The release from the stainless-steel

Table 6.9. Fission Product Release[a] from Trace-Irradiated PWR-Type UO_2 Melted in a Single Element Tungsten-Resistor Furnace Filled with Helium[b]

Element	Heat Duration (min.)	UO_2 Vaporized (%)	Gross γ Release (%)	Percent of Individual Fission Product Released							
				Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Ce/RE
UO_2	5.0	0.8	7.1	63	47	56	44	1.6	1.6	5.3	<0.6
UO_2	4.0	0.2	5.7	50	30	42	41	0.4	0.8	2.9	<0.5
UO_2	4.4	0.3	6.9	34	25	33	<40	0.05	1.2	4.3	0.5
UO_2 (SS clad)	4.7	0.2	5.0	56	52	31	46	0.5	1.0	4.2	0.3
UO_2 (Zr clad)	7.0	0.1	2.6	52	24	1.1	28	0.1	10.1	10.6	0.5
UO_2 (Zr clad)	6.7	0.04	5.2	41	50	0.6	32	0.2	10.0	7.5	0.5

[a] Results are not corrected for the fraction of the sample melted which is approximately equal to the percent rare gas release. Release is from fuel and cladding.

[b] Helium flow rate, 400 cc/min.

clad specimen was similar to that from unclad fuel but the zirconium clad specimens gave quite different results. The data in Table 6.9, coupled with post-melting examinations, indicate that the molten zirconium wet the UO_2 and spread over the surface. It thus served as an effective oxygen-getter, which accounts for the high strontium and barium escape. Cesium, iodine, and rare gases were apparently unaffected by the cladding.

Fission-product release experiments[62] in the Containment Mockup Facility (CMF) located in a hot cell were performed with a 25-kw, 5-Mc induction heater (Fig. 3.11). Stainless-steel-clad UO_2 specimens supported in zirconia crucibles and surrounded by a quartz furnace tube are heated by induction coupling to the cladding material. By the time the cladding melts, the UO_2 is hot enough to become conducting and direct coupling by the high-frequency generator is accomplished to heat the UO_2 to its melting point. The duration of the period in the molten state was limited by the time required for the molten UO_2 to penetrate the zirconia crucible wall (approximately one minute). Results obtained with two tracer-level-irradiated fuel specimens and one irradiated to 7000 Mwd/ton are displayed in Table 6.11.

Table 6.11. Release of Fission Products From Stainless-Steel-Clad UO_2(a) Melted in Air by Direct Coupling Induction Heating

Total Release (%)											
Iodine			Tellurium			Cesium			Plutonium		
A	B	C	A	B	C	A	B	C	A	B	C
84	64		12	12	55	29	37	37			0.005

---

[a] Runs A and B were made with tracer-level-irradiated UO_2. Run C was made with 20g of UO_2 irradiated to 7000 Mwd/ton.

A comparison of fission-product release results obtained by different methods of melting UO_2 is given in Table 6.12. It is apparent that a large fraction of the rare gases, iodine, tellurium, and cesium will be released when UO_2 melts, except that tellurium may be retained by molten zirconium cladding. Ruthenium release was large only in the arc-image furnace experiments and, to a much lesser extent, in the ORR in-pile experiments.[63] In the former case, only ceramic materials (BeO and UO_2) were present in the high-temperature zone and consequently oxygen liberated from the UO_2 specimen may have contributed to the volatility of the ruthenium rather than being adsorbed by hot tungsten or cladding material. The release of strontium and barium was high only in the experiments with center resistor-zirconium clad specimens (oxygen getting action of the cladding material).

6.2 Uranium Oxide-Beryllium Oxide

The excellent nuclear and physical properties of BeO make it attractive for use as a diluent for uranium dioxide in ceramic fuel elements.

Measurements of fission-product release accompanying the melting of fuel specimens of this type have been reported by Conn et al.[64] and by Parker and co-workers.[40]

A more thorough investigation of parameters affecting the release of fission products from small tubular samples of BeO-UO_2-Y_2O_3 fuel was made[40] by use of the arc-image furnace (Fig. 3.5). The fuel specimens were surrounded by a glass envelope during fission-product release experiments (see Fig. 3.6). Air flowing through this envelope carried particles and gases evolved from the heated fuel to the collection train (Fig. 3.6). The length of time molten was not well controlled with this heating arrangement. The tubular fuel specimens were held in a horizontal position with the image of the carbon arc focused on its front end initially.

Table 6.12. A Comparison of Fission-Product Release by Different Methods of Melting UO_2 in Helium

Method of Melting	Sample Weight (grams)	Percent UO_2 Vaporized	Gross γ	Percent Release							
				Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Ce
Arc-image[a]	0.25	~2.0	~30	99.5	95.2	96.2	92.9	78.3	3.9	7.3	3.8
Tungsten crucible	29.0	0.16	15.1	98.0	98.3	97.6	66.0	0.05	0.47	2.57	0.07
Tungsten resistor (unclad)[b]	39	0.8	14	~100	70	90	82	1.1	2.5	9.0	1.0
Tungsten resistor (SS clad)[c]	39	0.4	9	~100	93	55	83	0.9	1.7	7.4	0.5
Tungsten resistor (Zr clad)[d]	39	0.17	8	~100	83	1.8	66	0.4	22	20	1.2
In-pile (ORR)[e]	6	1			90	75	77	4	1.5	1	0.3
In-pile (TREAT)[f]	30	0.01			5.4	0.8	3.8	0.03	3.2	2.2	0.01

Induction

[a] Impure helium atmosphere.

[b] Average of three runs, data normalized to 100% Xe-Kr release.

[c] Actual data normalized to 100% Xe-Kr release. The numbers of release from fuel and cladding. The retained Te was equally divided between cladding and fuel.

[d] Average of two runs, data normalized to 100% Xe-Kr release.

[e] Release from high-temperature zone.

[f] Release from high-temperature zone during ~0.1 sec transient.

When this part of the specimen melted, the molten portion dropped far enough to be out of the high-temperature zone and the specimen was advanced so that another portion was heated to the melting point. This was continued until all the specimen and a small part of the solid beryllium oxide rod used to support the specimen were melted. The time required to complete the operation varied from about 80 to 95 seconds. Data obtained with three types of fuel are recorded in Table 6.13. Most of the melting experiments were performed with fuel irradiated to 0.43% burnup of ^{235}U but two experiments were made with low-burnup (0.01%) material. Most of the specimens were re-irradiated to build up an inventory of short-lived isotopes.

High release values for iodine, tellurium, cesium, and ruthenium are noted in Table 6.13 and a significant fraction of the uranium content of the fuel and a smaller amount of the BeO also volatilized. This enhanced volatility of both uranium and beryllium could have been an effect of the high surface/volume ratio of the very small samples. No significant burnup effect was shown over this narrow range of burnup and a 10-fold increase in air velocity likewise had little effect on fission product release although it had a large effect on fission product transport. Maximum fuel melting temperatures measured by means of an optical pyrometer in these experiments were 2550 ± 30°C and freezing temperatures of 2450 ± 25°C were recorded. These values are somewhat higher than the value reported elsewhere[61] (2315°C) for this type of fuel.

Some data were also obtained on the rate of release of fission products from ceramic coated fuel elements heated for 5-hr periods in flowing helium. The data are presented graphically in Fig. 6.11 in a form that permits extrapolation to temperature outside the measurement range (1015 to 1400°C).

Table 6.13. Release of Fission Products from $BeO-UO_2-Y_2O_3$ Fuel Specimens - Melted in Air

Fuel Type	Rare Gases	Gross γ	Percent of Total Activity or Fuel Component Vaporized										
			I	Te	Cs	Ru	Sr	Zr	Ba	Ce	TRE	Be	U
Uncoated	75.7	11.6	56.2	62.7	61.5	55.8	0.03	0.02	0.3	0.6	0.26	0.86	8.9
Uncoated	71.3	11	76.7	68.2	62.0	63.7	0.15	0.02	0.54	0.5	0.19	0.58	8.2
Uncoated	77.1	11.6	79.0	75.1	50.1	64.4	0.11	0.03	0.56	0.4	0.45	1.6	7.8
Uncoated[a]	73.8	7.6	70.7	66.3	45.7	54.2	0.02	0.42	0.22	0.5	0.18	0.76	9.3
Uncoated	82.0	10.0	83.4	84.5	41.4	61.9	0.11	0.01	0.13	0.4	0.12	0.58	16.1
Uncoated	69.7	26.0[b]	72.4	71.0	72.9	66.9	0.10	0.02	0.39	0.6	1.4	0.89	12.6
Uncoated[c]	69.5	18.4[b]	78.8	71.7	49.3	57.6	0.05	0.03	0.15	0.5	0.28	1.23	7.8
Coated[e]	53.4	10.6[b]	73.5	63.2	44.6	50.0	0.08	0.004	0.5	0.9	0.19	0.60	9.8
Coated	57.0	13.8[b]	77.3	72.4	32.3	58.3	0.02	0.005	0.5	0.42	0.29	0.69	10.6
Coated	d	4.9	d	64.3	60.9	49.9	0.02	0.009	0.16	0.48	0.27	0.66	13.0
Inside	59.4	3.87	73.5	68.4	33.5	48.3	0.008	0.0034	0.156	0.072	0.2	0.327	5.91
Only	59.2	5.8	66.8	76.6	55.4	55.3	0.013	0.005	0.15	0.18	0.14	0.339	5.76
Coated[e]	60.9	5.7	63.3	78.7	57.6	59.6	0.01	0.004	0.23	0.45	0.2	0.366	6.66

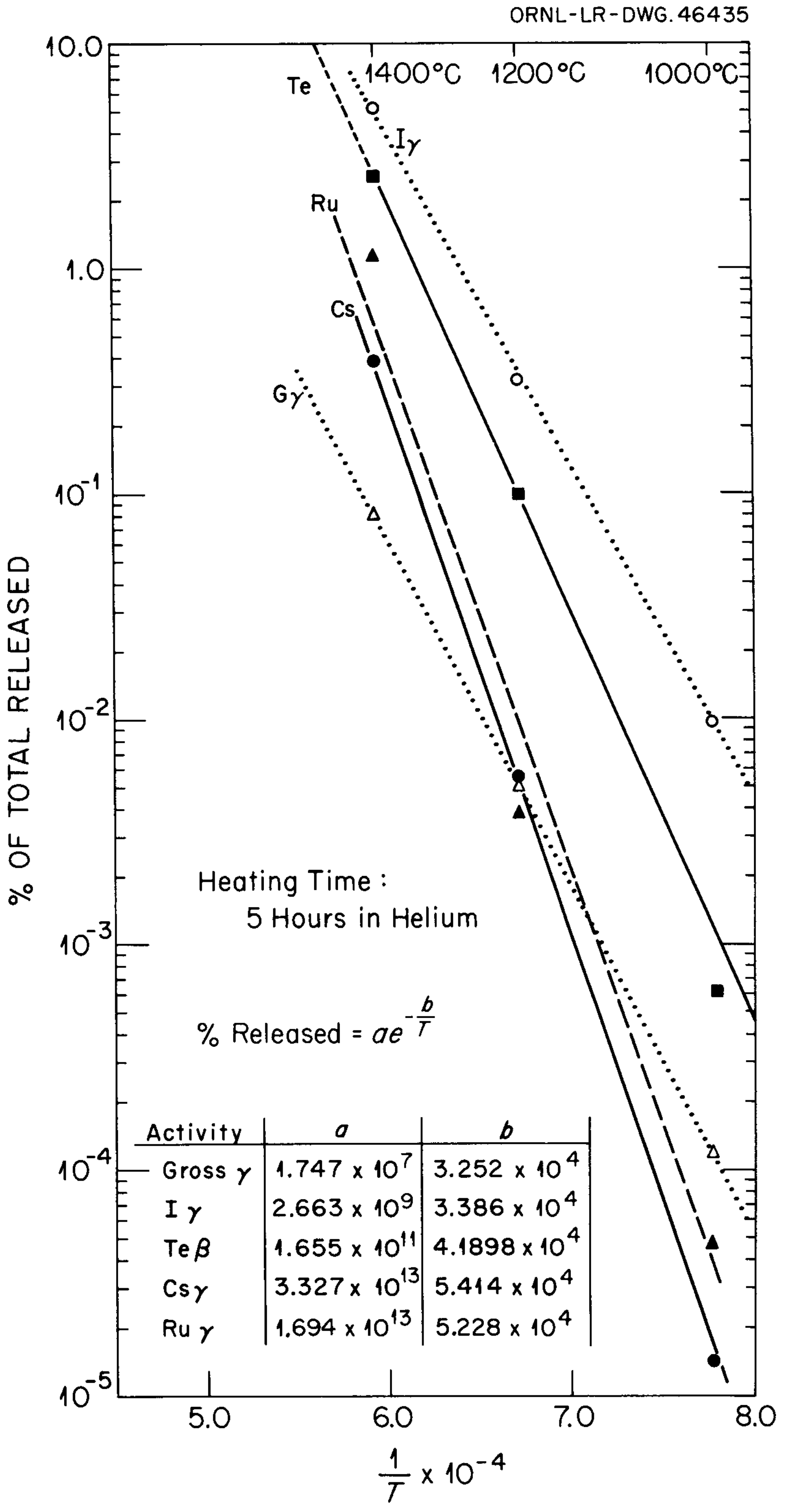
[a] 0.01% burnup of ^{235}U. All other specimens irradiated to 0.43% burnup.

[b] High release of gross gamma due to short cooling period after re-irradiation, resulting in presence of more volatile short-lived gamma emitters.

[c] Air flow velocity in this experiment was 5 cfm. In all other experiments it was 0.5 cfm.

[d] Not re-irradiated.

[e] Coatings consisted of a few mils of ZrO_2 or pure BeO.



Activity	a	b
Gross γ	1.747×10^7	3.252×10^4
I γ	2.663×10^9	3.386×10^4
Te β	1.655×10^{11}	4.1898×10^4
Cs γ	3.327×10^{13}	5.414×10^4
Ru γ	1.694×10^{13}	5.228×10^4

Fig. 6.11. Release of Fission Products on Heating Ceramic-Coated BeO-UO_2-Y_2O_3 Fuel for Five Hours in Flowing Helium.

7.0 SUMMARY AND DISCUSSION

Various types of reactor fuel materials have been of sufficient interest in different AEC programs during the period covered by this report to warrant investigation of their fission product release characteristics. We have not exhausted the list of possible fuels by any means and only a cursory study of fission product release from some fuels was made. Nevertheless a fairly representative group of fuels is covered by this report. The principal exceptions are graphite fuels which are considered only in Section 2. In the earlier years of this study, uranium and uranium alloys received major attention and the amount of space devoted in this report to such fuels, especially pure uranium, is out of proportion to their current importance. Since 1960, UO_2 fuels have assumed major importance in the power reactor field and this type of fuel has been investigated thoroughly in this program and elsewhere and it continues to be the "standard" fuel for water reactor investigations.

Oxidation and melting are the principal mechanisms that result in fission product release from overheated uranium. Uranium alloys such as U-Al and U-Zr oxidize less readily than pure uranium, in general, but their low melting points make melting the most likely mode of release. The more volatile fission products including rare gases, iodine, tellurium, and cesium are the only species released to any extent when such alloys melt.

Diffusion, as well as oxidation on melting, is an important mechanism of release from UO_2 fuels because there is a large temperature range (approximately 1600 to 2800°C) in which UO_2 neither melts nor oxidizes. The lower end of this range is above the melting point of stainless steel cladding and the other common cladding material, Zircaloy, melts at about 1850°C. Diffusion rates are accelerated by UO_2 grain growth which is a function of both time and temperature but this effect becomes significant at around 1700°C. Prompt burst release, which occurs when fission products that diffuse from the fuel into cladding void space escape on failure of the cladding, is a release mechanism that remains to be investigated. Oxidation of UO_2 after the de-cladded material cools below 1550°C should not be overlooked as a potential cause of fission product release.

8. THE APPLICATION OF FISSION PRODUCT RELEASE DATA TO REACTOR HAZARDS ANALYSES

A detailed treatment of the application of fission product release data to reactor hazards analyses is beyond the scope of this paper but, in order to place the numerical values of percent of fission products released from overheated reactor fuels in proper perspective, it is perhaps worth while to examine the mechanics of the process of formulating a hazards analysis. In its simplest form there are three major subdivisions of the analysis each of which requires the best possible evaluation of the numerical data that apply specifically to the particular reactor under consideration and to its characteristics and environment. These subdivisions are:

I. The fission-product source term, that is, the amount of radioactivity leaving the reactor core and reaching the containment barrier, calculated with proper credit for natural deposition processes and engineered safeguards.

II. The rate of containment leakage to the atmosphere with credit for leakage reduction that results from diminishing pressure.

III. The radiological hazard presented by the radiation source (gamma activity from fission products within the containment shell) and the atmospheric dispersion of radioactivity that escapes from the containment system.

In this report, we have confined our attention to the first part of I, the determination of the amount of radioactivity leaving the fuel.

The AEC Division of License and Regulation in TID-14844, and in the Federal Register (Part 10CFR100), suggests typical (not actual) values for civilian power reactors for the source term as applied to UO_2-fueled water reactors:

1. 100% of the rare gases (8% of total gamma after 1 day)
2. 50% of the halogens (4.5% of total gamma after 1 day)
3. 1% of solids (rare earths, and refractory elements, Zr, Nb, Ba, Sr, etc.) (0.5% of total gamma after 1 day).

Growth in the number and size of power reactors has resulted in increased emphasis on the use of specific values rather than typical values. In fact, while the biological significance of other fission products may not be so pronounced, the conclusion obtained from most fission product release (threshold melting of UO_2) experiments suggests that the total quantity of released fission products should include in addition to the above:

a. 50% of cesium (0.05% of total gamma after 1 day).

b. 50% of tellurium (oxidizing atmosphere)(2% of total gamma after 1 day).

c. 5% of ruthenium (~ 1% of total gamma after 1 day) and

d. 0.01% of plutonium.

Calculation of the fission product source term is especially difficult when the hazards analysis indicates that the core may be only partly melted with some regions undergoing lesser amounts of damage and the melted portion being redistributed to a configuration that permits it to remain at a high temperature for a significant length of time.

Under such circumstances it is necessary to consider the accident to be a series of separate chronological events contributing to fission product release from each region of the core. These include:

1. Prompt release on clad rupture (below 1500°C for SS clad fuel; 1850°C for Zircaloy cladding).
2. Release by high temperature diffusion and grain growth.
3. Release and/or retention by cladding interaction.
4. Release by melting and eutectic formation.
5. Release by oxidation on cooling.

The summation of the fission product release resulting from each of these processes is necessary to obtain a final release value; however, it is also necessary to make such a summation on a chronological basis in order to apply the proper containment leakage and atmospheric dispersion values.

If one estimates the fraction of total gamma energy in the volatile fission products, this value may be as much as 15 to 20%; however the transport process from the primary vessel is dependent on many variables such as gas displacement, the oxidizing or reducing nature of the fuel environment, maximum metal surface temperatures, reassembly of fuel for reheating, so that the amount of volatile fission products (except the rare gases) actually reaching the containment shell should be at least an order of magnitude less than the amount volatilized.

It should be clear from this discussion that it is much more important to know an approximate value of the transport efficiency of certain fission products than it is to know the precise values of release from the fuel.

Since the advent of better engineered safeguards, it is possible, too, that the worst accident condition a reactor core will ever experience will be cladding rupture at blowdown. In-core spray cooling prevents further overheating. In this case, the prompt release of fission products from the fuel void space, which varies mainly with operating heat rating (center-line fuel temperature) and burnup, would be the controlling source term.

Engineered safeguards such as sprays and filters have improved in reliability so that the reactive forms of iodine and the airborne solid particles are rapidly removed from the containment atmosphere. This results in the limiting hazard condition becoming the amount of iodine converted to the organic (methyl iodide) form and the amount of rare gases.

The biological significance of the fission products as an ingestion or inhalation hazard has been rated by Beattie and others in the following order:

1. Iodine (most important).
2. Tellurium, 1/10 as significant as iodine.
3. Strontium and cesium, 1/10 as significant as tellurium.

There is no safeguard for removing rare gases available at the present time but high-pressure storage, barrier diffusion, and isotopic exchange offer possible solutions to this problem.

Since the practical value of safeguards is now being demonstrated, credit for reduction of fission products by:

1. Removal of airborne fission products by spray and filtration systems,
2. Removal of airborne fission products by deposition in containment vessel, and
3. Removal of pressure by suppression devices (cooling or pool suppression),

may be of such value that a reduction by more than two orders of magnitude in the amount of airborne fission products can be expected. At a maximum containment leakage rate of 0.1% per day, assumption of perhaps two or three more orders of reduction in the amount of fission products escaping to the environment would be reasonable.

In order to promote confidence in such large reduction factors, continued research into the efficiency of removal for all the various forms of the released fission products will be required.

In conclusion, we wish to emphasize that there are many factors affecting the fission product source term and the amount of fission products which actually can escape the containment system of power reactors in reactor accidents. While the amount of fission products evolved from overheated fuel is highly useful information, it is now recognized that the hazard of reactor accidents can be fully evaluated only through sophisticated accident simulation experiments in facilities such as the Containment Research Installation (ORNL), the Containment Systems Experiment (Battelle Northwest), and the Loss-of-Fluid Test (Phillips-Idaho).

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