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A REVIEW OF THE U_3O_8-ALUMINUM REACTION AS A POTENTIAL HEAT SOURCE IN RESEARCH AND TEST REACTOR ACCIDENTS*

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ABSTRACT

A critical review of the literature on the U_3O_8-aluminum reaction has been conducted. The reaction in fabricated fuel plates is found to be less energetic and much slower than in cold-pressed powder mixtures. The difference is at least partially attributable to conversion of up to 50% of the U_3O_8 to U_4O_9 during fabrication. No definitive measurements of the amount and rate of energy release have been made. Data are provided upon which to base calculations of energy release.

INTRODUCTION

Since the discovery in the late 1950s that U_3O_8 reacts less rapidly with aluminum at fabrication temperatures than does UO_2, U_3O_8-Al dispersion fuel has been adopted by a number of reactors, e.g., HFIR, HFBR, NBSR, RA-3 (Argentina) and the 30-MW Multipurpose Reactor (Indonesia). Fuel elements of the NBSR, HFBR, and HFIR contain from ~33 wt% to ~45 wt% of 93%-enriched U_3O_8 in the fuel meat, while the fuel elements of the two foreign reactors contain from 65 to 75 wt% of 19.75%-enriched U_3O_8. Consideration has been given to using U_3O_8-Al dispersion fuel containing up to 62 wt% U_3O_8 in the production reactors at the Savannah River Site.

It was recognized very early that a mixture of U_3O_8 and aluminum is not in chemical equilibrium and that the potential for a significant exothermic reaction existed. The earliest experiments, at Georgia Tech (GT),[1,2,3] demonstrated that mixtures of U_3O_8 and Al powders could react violently, and the term "thermite" was later attached to the reaction. Further studies at Argonne National Laboratory (ANL) in the 1960s,[4,5] Oak Ridge National Laboratory (ORNL) in the late 1970s,[6] and Savannah River Laboratory (SRL) in the 1980s[7,8,9] have successively provided significant additional information but, unfortunately, have not provided a complete understanding of the reaction. The resulting data must be carefully interpreted to evaluate the potential of the U_3O_8-Al reaction as a important heat source in reactor accidents. This paper provides an overview and interpretation of the available data.

The primary issues addressed in this review are the nature of the reaction, the onset temperature, the amount and rate of energy release, the potential for self-propagation, and the effects of fuel burnup on the foregoing. First, the types of experiments performed will be summarized; then the issues listed above will be discussed.

SUMMARY OF EXPERIMENT TYPES

Samples

A dispersion fuel plate (or tube) is produced by cold pressing a mixture of fuel powder and aluminum powder to produce a compact, enclosing the compact inside a suitable aluminum alloy clad to form a billet, and hot rolling (or hot extruding) the billet at temperatures between 425 and 525°C to produce the fuel plate (or tube). The process can include a number of annealing steps in the same temperature range to degas the billet prior to rolling and to test the quality of the plate (blister testing). The fuel meat is typically 0.5 to 1.25 mm thick and, for U_3O_8, contains from 12 to 45 vol% (30 to 75 wt%) fuel. The average fuel powder particle size is typically of the order of 75 µm with an upper limit of 125 to 150 µm and from 15 to 40 wt% <44 µm. In general, the aluminum powder is considerably finer.

Because hot rolling and annealing promotes intimate contact and chemical reactions between the U_3O_8 and Al particles, the conditions inside the fuel meat of a fuel plate may be considerably different than in a cold-pressed mixture of U_3O_8 and Al. Nevertheless, because of the ease of fabrication of samples, most of the specimens for the exothermic reaction tests (especially those for parametric studies) have been produced by cold pressing. The tests at GT, ANL, and SRL used cold-pressed pellets containing from 30 to 90 wt% U_3O_8 and ranging from 9.5 to 25.4 mm in diameter and from 12.7 to 25.4 mm in length. The sample volumes ranged from ~1 to 13 cm^3 and contained of the order of grams or tens of grams of U_3O_8. Some of the tests at ORNL and SRL made use of much smaller samples, crushed from cold-pressed pellets, which contained only tens of milligrams of U_3O_8.

Tests with more prototypic samples have been performed at ORNL using small samples punched from fabricated miniature fuel plates and at ORNL and SRL using either miniature fuel plates (~50 x 115 mm) or sections cut from full-sized fuel tubes. The fuel loadings studied were 75 wt% U_3O_8 at ORNL and 53 wt% U_3O_8 at SRL. However, when one includes the aluminum of the cladding of the fuel plate or tube specimens, the U_3O_8 weight fraction is considerably reduced--e.g., to 33 to 44 wt% for the ORNL samples.

Measurement Techniques

In general, two techniques have been employed: heating in a furnace with temperature measurements by thermocouple or optical pyrometry or with visual observation without temperature measurements, and differential thermal analysis (DTA). Furnace heating rates have varied from tens to hundreds of degrees Celsius per minute. Heating rates in the DTA experiments ranged between 10 and 33°C/min. The most accurate determinations of onset temperature came from the DTA experiments because compensation for the heating effect of the furnace is provided automatically by simultaneous heating of a reference sample.

The groups at GT, ORNL, and SRL used x-ray diffraction (XRD) to identify the reaction products. The GT group also used electron microprobe analysis, and the ANL group used quantitative chemistry.

DISCUSSION OF THE MAJOR ISSUES

Nature of the Reaction

Based on the results of XRD studies of the reaction products, the GT group[3] concluded that the reaction occurred in two stages: first the relatively slow reduction of U_3O_8 to UO_2, beginning at 649°C, followed by the rapid reduction of UO_2 to one or more of the uranium aluminides, beginning between 816 and 1066°C, according to the following reaction equations:

$$U_3O_8 + 4/3\ Al \longrightarrow 3\ UO_2 + 2/3\ Al_2O_3 \qquad (1)$$

$$UO_2 + (4/3 + x)\ Al \longrightarrow UAl_x + 2/3\ Al_2O_3, \qquad (2)$$

where UAl_x represents one or more of the uranium aluminides (UAl_2, UAl_3, or UAl_4). They found the reaction products to be α-Al_2O_3, UO_2, UAl_2, and UAl_3 in high-loaded U_3O_8 dispersions and α-Al_2O_3, UO_2, UAl_3, and UAl_4 in low-loaded U_3O_8 dispersions. No quantitative determination of the amounts of these reaction products was made. No residual U_3O_8 or Al were found.

The ANL data are basically consistent with the GT data--oxides and aluminides were found, with about equal amounts of UO_2 and Al_2O_3.

ORNL used XRD to determine the reaction products present after DTA runs using cold-pressed samples containing 50 wt% U_3O_8 and reaching, successively, 725, 1250, and 1350°C. They found U_3O_8 remaining after each run, indicating incomplete reaction. Al_2O_3 appeared after the 1250°C run and UO_2 and UAl_2 after the 1350°C run. These qualitative data are, again, consistent with Eqs. (1) and (2), although it is somewhat surprising that no UAl_3 was seen. Apparently, the heat generation rate was low enough and, perhaps, the sample size was small enough (tens of milligrams of U_3O_8 compared to a few grams in the GT experiments) that it either did not reach, or remain long enough above, the temperature needed to complete the reaction.

The SRL group[7,9] recently made the significant determination, based on XRD, that a substantial amount of U_3O_8 is converted to U_4O_9 during the fabrication process, according to the equation:

$$U_3O_8 + 5/6\ Al \longrightarrow 3/4\ U_4O_9 + 5/12\ Al_2O_3. \qquad (3)$$

The U_4O_9 can subsequently react with the remaining Al according to the equation:

$$U_4O_9 + 2/3\ Al \longrightarrow 4\ UO_2 + 1/3\ Al_2O_3. \qquad (4)$$

They found that ~50% of the U_3O_8 was converted to U_4O_9 during the Savannah River fabrication process. The effects of processing temperature and processing time on the conversion rate are shown in Fig. 1.

The SRL group is confident in its identification of U_4O_9 as the product of reaction (3)[10]. However, one must then question why U_4O_9 was not identified in previous studies. The explanation is most likely that for many years there was some question as to the existence of U_4O_9 as a distinct phase and that the published XRD pattern for U_4O_9 is very similar to that for UO_2.[11]

Unless one were specifically looking for U_4O_9, it would be very easy to mistake it for UO_2 in an XRD analysis. In addition, XRD analyses in the earlier experiments had been performed on cold-pressed compact samples, which had not experienced the temperature-time histories of fabricated plates and, therefore, contained relatively little U_4O_9.

The Al_2O_3 produced in either reaction (1) or (3) would be expected to form at the interface between the U_3O_8 particle and the Al, where it would act as a diffusion barrier, slowing down further reaction. As will be discussed below, this barrier may play a significant role in determining the onset temperature of the exothermic reaction.

When studying the results of experiments on the U_3O_8-Al reaction it quickly becomes evident that the mechanism is complex due to the variety of possible reactions and to the chemical and physical states brought about by these reactions. The U_3O_8 begins to react during fabrication and the extent of that reaction is determined by the temperature-time history during fabrication. The best evidence to date is that Eq. (3) governs the reaction during fabrication. During further heating, more U_4O_9 may be produced and perhaps some UO_2 according to Eq. (1). The experiments indicate that a rapid reaction of U_4O_9 and or UO_2 with Al occurs during one temperature range, governed by a combination of Eqs. (4) and (2) and that a rapid reaction of residual U_3O_8 occurs at a higher temperature, governed by a combination of Eqs. (1) and (2).

Onset Temperatures

As stated previously, the most accurate determinations of onset temperature have come from the DTA experiments. In experiments with cold-pressed compacts the first exotherm consistently appears 20-40°C below the melting point of Al. It is completely dominated by the Al-melting endotherm. The GT group attributed this to reaction (1), but it now seems that, more likely, it is attributable to reaction (3). This reaction begins at lower temperatures, but there it is a diffusion-controlled solid state reaction and, hence, too slow to be detected by DTA. Nearer the melting temperature of Al, however, it is likely that some of the aluminum close to the reaction front becomes molten and the rate of reaction significantly increases. This low-temperature exotherm is not seen in DTA studies using samples from fabricated plates because a significant amount of the reaction would have already occurred during fabrication, leaving a barrier of reaction products around the remaining U_3O_8.

The next exotherm occurs at a temperature between 850 and 1000°C, although the GT group reported that some samples reacted at 816°C. Within this broad agreement, there is some inconsistency among the results of the different groups. The onset temperature fell between 927 and 982°C in most of the GT experiments, which used cold-pressed compacts; between 850 and 880°C in the ORNL experiments with cold-pressed compacts; and between 872 and 908°C for the ORNL experiments with plate samples. The SRL group found, using compacts, that the major energy release begins at about 950°C, although there is some evidence for a small reaction at about 850°C.[12] The GT group found a distinct effect of U_3O_8 particle size, with the onset temperature increasing to as high as 1066°C for particles of size >149 μm, and of U_3O_8 loading, with the onset temperature increasing with loading in the range studied, 74.5 to 85.4 wt% U_3O_8.

The ORNL group found one or more additional exotherms with onset temperatures in the 1150 to 1250°C range in many, but not all, tests using both cold-pressed compacts and fuel plate samples. These high-temperature exotherms were well-separated in time (by tens of minutes) from the exotherm occurring near 900°C. The source of these reactions was not identified. No such reactions have been reported in other DTA experiments, either because sample temperatures were too low or because the samples reached these temperatures so rapidly that the two reactions were indistinguishable in time. The temperature-time traces from furnace experiments at both ANL[5] and SRL[7,8] do show a second exotherm with an onset temperature of about 1300°C. In the ANL experiments, they occurred two to three minutes after the initial reaction.

The SRL group carried out an extensive set of DTA experiments using compacts which had been annealed for times needed to convert various amounts of U_3O_8 to U_4O_9.[9] The results of their study are shown in Fig. 2. The SRL group has inferred from these data that the onset temperature of the U_4O_9-Al reaction is in the 850 to 900°C range and the onset temperature of the U_3O_8-Al reaction is in the 1150 to 1200°C range. The latter conclusion appears to be well justified by the data; however, a number of samples containing very large amounts of U_4O_9 reacted in the 1100 to 1200°C range. Therefore, the amount of U_4O_9 in the sample may not be the independent variable.

In fact, it has been suggested that polymorphic phase transitions in Al_2O_3 may play a significant role in initiating the exothermic reaction.[13] It does seem more than a coincidence that Al_2O_3 phase changes, accompanied in some cases by very large volume changes, occur at about the temperatures where exotherms have been observed (e.g., $\gamma \rightarrow \theta$ at 850-900°C, $\delta \rightarrow \alpha$ at 950°C, and $\theta \rightarrow \alpha$ at 1100-1150°C) and that Al_2O_3 is wetted by Al at 940 and 1255°C.[14] It appears likely, therefore, that the Al_2O_3 barrier cracks and allows molten aluminum to flow to the reaction front in sufficient quantity to sustain a rapid reaction. The actual temperature at which the barrier is broken might well depend on the thickness of the barrier and, hence, on the temperature-time history of the dispersion. For example, a sample heated long enough to produce a significant amount of U_4O_9 might have a thicker coating of Al_2O_3 around the fuel particles, which would be more likely to crack during a phase transformation, leading to an earlier (lower-temperature) onset of the reaction. The fabrication process (rolling or extrusion) can affect the Al_2O_3 barrier by introducing cracks or residual stresses which might contribute to earlier cracking upon subsequent heating.

The onset temperature data illustrate the complexity of the U_3O_8-Al reaction. In most cases there is an exotherm with an onset temperature between 850 and 950°C, most likely from a reaction of U_4O_9 and/or UO_2 with Al. The onset temperature appears to be influenced by the powder and fabrication variables and the temperature-time history of the sample. In many, but not all, experiments higher-temperature exotherms were found, some beginning about 1100°C and some beginning about 1250°C. The SRL experiments indicate that the higher of these is produced by the reaction of residual U_3O_8 with Al.

Amount and Rate of Energy Release

The amount and rate of energy release during the exothermic reaction has exhibited considerable variation among the experiments. As well as resulting from the use of different types of samples--cold-pressed compacts and pieces of fabricated fuel plates--mentioned earlier, some of this variation undoubtedly resulted from the use of greatly different sample sizes.

The GT group observed what were described as "violent" reactions in cold-pressed compacts, where they measured surface temperatures, by optical pyrometry, as high as 2300°C and temperature rises of thousands of degrees Celsius per second. Relative peak areas from their DTA measurements indicated that the energy release increased as the U_3O_8 content decreased from 85.4 to 74.5 wt%. They found that the energy release was a function of U_3O_8 particle size, peaking in the 53-74 µm size range. The ANL group found peak temperatures and heating rates in compacts containing 85 wt% U_3O_8 comparable to those observed by the GT group only when the compacts had been sintered in air at 600°C for 4 h prior to performing the experiment. They concluded that the slow heating rate employed by the GT group might have served to presinter their samples. The SRL group observed the specimen to glow white hot but did not observe any disruptive reactions. All of these measurements were performed on compacts ranging in volume from ~1 cm^3 at GT to almost 14 cm^3 at SRL. Perhaps the SRL sample size was so large that the reaction was spread over time throughout the pellet, leading to a smaller temperature rise. Using the temperature-time graphs, the SRL group estimated that the energy released from the 53 wt% U_3O_8 pellets was ~820 kJ/mole of U_3O_8.

The ORNL group made a careful quantitative measurement of the energy released in the exotherm occurring near 900°C. The energy released in the higher-temperature exotherms could not be measured because their equipment was calibrated only up to ~1100°C. Their result for the cold-pressed compact sample containing 79 wt% U_3O_8 was 85 ± 5 kJ/mole of U_3O_8. Their XRD measurements indicated that a significant amount of the U_3O_8 had not been converted either to U_4O_9 or UO_2, so that much of the energy would be expected to be released in the high-temperature exotherms, in agreement with their published thermogram. Their results for the fuel plate samples were 461 ± 239 J/mole of U_3O_8 and 182 ± 46 J/mole of U_3O_8 for fuel/cladding mixtures containing ~44 wt% U_3O_8 and ~33 wt% U_3O_8, respectively. Again, the thermograms indicate that a considerable amount of energy might have been released in the high-temperature exotherm, especially for the 33 wt% sample. Individual measurements ranged from 237 to 672 J/mole of U_3O_8 for the samples containing ~44 wt% U_3O_8. Also, on a molar basis one would expect the two sets of measurements to give the same result. These variations do, indeed, appear to be real, are not understood, and again indicate the complexity of the reaction.

Both the ORNL and SRL groups performed experiments in which either whole miniplates or similar-sized samples cut from fuel tubes were heated in furnaces, at both slow or rapid rates. Peak furnace temperatures of 1405 and 1375°C, respectively, were reached. Although exothermic reactions were detected, they were rather small in magnitude and showed no major disruptive effects.

Although they made no absolute measurements, the GT group did calculate the maximum heat which could be released by the combination of reactions (1) and (2) to be 1566 kJ/mole of U_3O_8; using the latest thermochemical data, this value is reduced to 1271 kJ/mole. The recommended values of the heats of formation of the constituent compounds of these reactions are listed in Table I.[13,15,16] The heats of reaction calculated for stoichiometric mixtures of the reactants in Eqs. (1) - (4) are listed in Table II. The room temperature values of the heats of formation have been used because they do not change much over the range of interest, thereby eliminating the need to decide which temperature to use.

Using the heats of reaction in Table II, one calculates that the maximum energy which could be released by the complete reaction of a mixture containing 53 wt% U_3O_8 is 1048 kJ/mole of U_3O_8. The value estimated by the SRL group from a temperature-time graph, 820 kJ/mole, is 78% of that value. Considering that some U_3O_8 would have been converted during the heatup, the measured value is reasonably close to the predicted value.

The complexity of the reactions and the dependence of both the onset temperature and the energy released on details of the fabrication and on the temperature-time history of the fuel makes it difficult to accurately estimate the amount of energy which will be released and the rate of energy release by the U_3O_8-Al reaction. Measurements on fuel plates or tubes indicate that the reaction rates are considerably slower than those measured in the early experiments with cold-pressed compacts (taking place in minutes rather than seconds or fractions of a second), but no definitive quantitative data are available.

One must rely on calculated values when estimating the amount of energy released during the U_3O_8-Al reaction. If a conservative lower limit of the amount of U_3O_8 converted to U_4O_9 during fabrication can be established, the maximum energy release can be reduced accordingly. Conservative estimates could also be made about the partition of the energy among the various exotherms. It would be most conservative, of course, to assume that all of the energy is released in an exotherm beginning at 850°C. Finally, one must estimate the rate of energy release.

Calculations performed for some high power reactors have shown that the maximum energy which could be contributed by the U_3O_8-Al reaction during an accident is small compared to either fission or fission product heating during the minutes the data indicate are needed to complete the reaction. Evaluation of the consequences of an added heat source from the U_3O_8-Al reaction must be made on a case-by-case basis, however.

Potential for Self-Propagation

One of the major concerns about an energetic reaction between the constituents of the fuel is that such a reaction might propagate to involve a substantial amount of fuel. The GT work with pellets and the ANL work with sintered pellets indicates that energy was indeed released rapidly enough in highly loaded, cold-pressed specimens to cause self-propagation of the reaction. Tests made on fuel plates or plate samples have given no indication that the reaction might be self-propagating. As stated above, experiments in which miniature fuel plates or sections of fuel tubes have been heated to high temperatures have shown no evidence of disruption indicative of a propagating reaction. In addition, the SRL group showed that neither heating a fuel tube section with an acetylene torch nor lowering a fuel tube section into 1000°C aluminum was sufficient to initiate a self-propagating reaction.[17]

It appears, therefore, that the rate of energy release by the exothermic reaction in U_3O_8-Al fuel plates is low enough that the heat generated can be dissipated quickly enough to preclude self-propagation of the reaction.

Effects of Fuel Irradiation (Burnup)

All of the experiments discussed above have been performed with unirradiated test samples. One must consider if irradiation of the fuel might produce any condition which might be worse than that in unirradiated fuel. Micrographic studies of irradiated U_3O_8-Al dispersion fuel have shown that the reaction rate of U_3O_8 is apparently enhanced by irradiation to the extent that highly burned U_3O_8 particles have been completely reacted.[18] Even though the reaction product was identified as UO_2,[19] the accuracy of the measurement was not great enough to distinguish between UO_2 and U_4O_9,[20] so either reaction (1) or (3) might be occurring during irradiation. In any case, the amount of U_3O_8 available to react during an accident and, hence, the potential energy release is steadily reduced during irradiation.

The consequences of the exothermic reaction could also be increased if fission products or their compounds resulted in a significant lowering of the onset temperature. Other compounds have been found to lower the onset temperature of the classic thermite (Fe_2O_3-Al) reaction,[21] and the heat liberated by a reaction of sodium uranate salts with aluminum at 350°C has been found to be sufficient to initiate the U_3O_8-Al reaction.[22] An extensive DTA study at SRL[12] showed that impurities (>5 wt%) of alkali metal chlorides and alkaline earth oxides cause only minor changes in the onset temperature of the U_3O_8-Al reaction. A study of the high-yield fission products has concluded that there is little chance that a harmful compound would be formed, either one that would react with the aluminum or one that would form a eutectic with the aluminum.[23]

Experimental evidence also points to a low probability of initiation below the aluminum melting temperature of an energetic exothermic reaction in U_3O_8-Al dispersion fuel. During the course of three decades of development of U_3O_8-Al dispersion fuels, countless postirradiation blister annealing tests and a significant number of fission product release tests have been performed at temperatures up to and exceeding the solidus temperature of the cladding. No mention has been found in any report of an event during these tests which might indicate that an energetic reaction occurred. On the other hand, since most of these tests were performed on reasonably highly irradiated fuel, one might argue that extensive reaction of the U_3O_8 has removed one of the potential reactants. However, the resulting UO_2 or U_4O_9 would still have been available to react.

It is concluded that substantial theoretical and experimental evidence exists that the onset temperature of the U_3O_8-Al reaction will not be significantly affected by irradiation of the fuel.

SUMMARY AND CONCLUSIONS

A careful review of the literature on the U_3O_8-Al reaction has shown that the reaction is complex and depends on a number of variables, including fuel powder characteristics, fabrication parameters, and the temperature-time history of the fuel plate. The number of exotherms occurring above the aluminum melting point and their onset temperatures are quite variable, but it is clear that the first exotherm does not occur below ~850°C in fuel plates or tubes, well above the melting temperature of the Al alloy cladding. Although no definitive measurements of the energy released in the complete reaction have been made, the data do indicate that the reaction occurs much more slowly and releases less energy in fabricated fuel plates than in the cold-pressed samples used for many of the early experiments. Evaluations of the potential contributions of the energy released by the U_3O_8-Al reaction to the consequences of an accident must be based on calculated energy releases. It does appear, however, that energy is released slowly enough to preclude self-propagation of the reaction in fuel plates or tubes. There is no evidence to suggest that irradiation of U_3O_8-Al dispersion fuel could result in a lowering of the onset temperature of the reaction.

This heat source should be considered when evaluating the consequences of severe accidents in reactors using U_3O_8-Al fuel. However, other heat sources are required to raise the fuel to the onset temperature of the energetic reactions, and, in general, it appears that the additional heat contributed by the U_3O_8-Al reaction will not significantly affect the outcome of the accident.

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Table I. Values of Heats of Formation Recommended for Calculation of Energy Release in U_3O_8-Al Reactions

Compound	Heat of Formation, J/mole
UO_2	-1085.0[a]
U_3O_8	-3574.8[a]
U_4O_9	-4512.0[a]
UAl_2	-92.5[b]
UAl_3	-108.4[b]
UAl_4	-124.7[b]
Al_2O_3	-1676.8[c]

[a] Ref. 15
[b] Ref. 16
[c] Ref. 14

Table II. *Heats of Reaction Calculated for Possible Stages of the* U_3O_8-Al *Reaction*

Reaction No.	Amount of U_xO_y,[a] wt%	Heat of Reaction,[b] kJ/mole of U_xO_y
3	97.4	507.9
4	98.4	386.9
1	95.9	798.1
2, x=2	73.4	125.4
2, x=3	68.5	141.3
2, x=4	64.3	157.6

[a] In stoichiometric mixture.
[b] Derived from heats of formation in Table I.

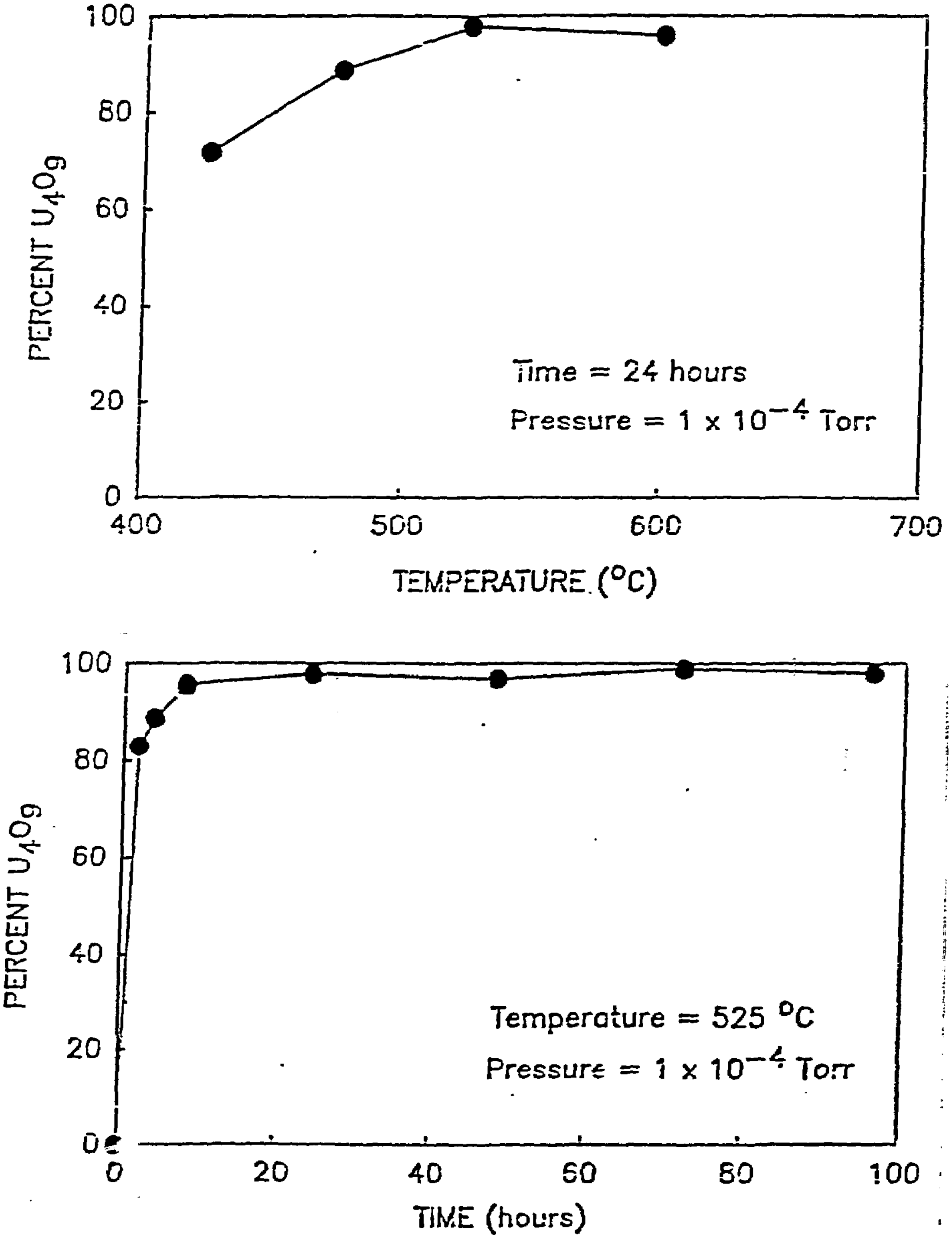

Fig. 1. Effects of Temperature and Time on the Amount of U_3O_8 Converted to U_4O_9 During Fabrication. (Ref. 9)

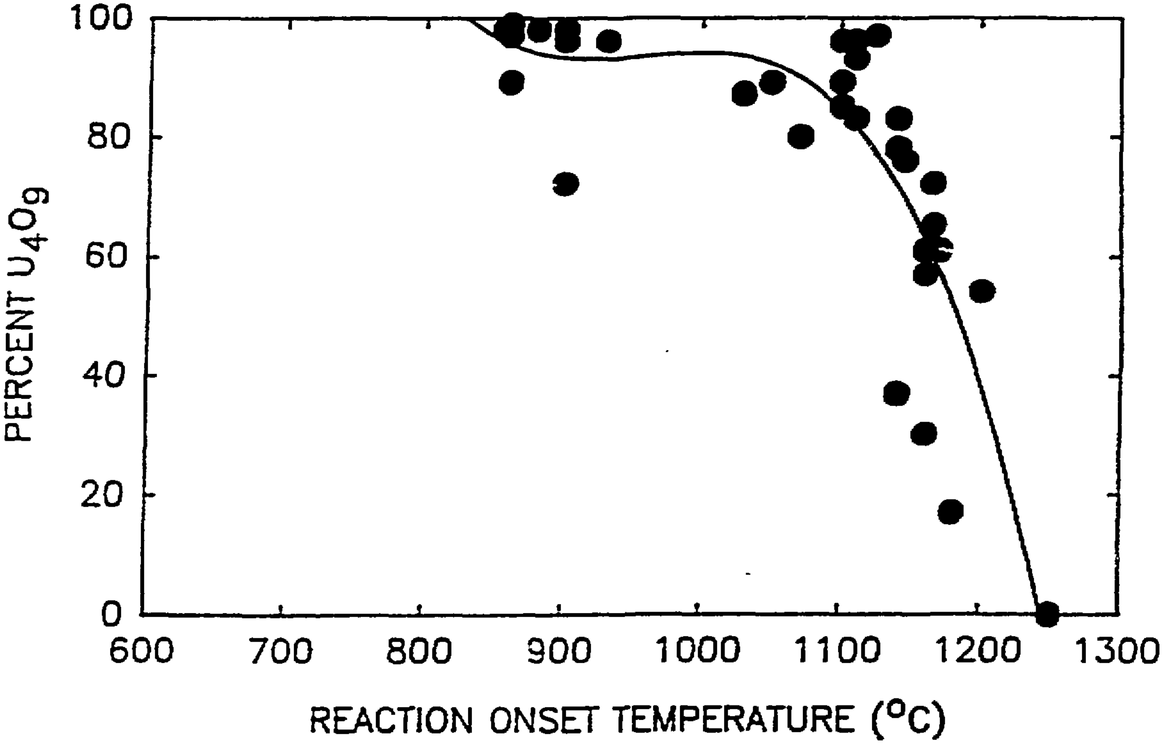

Fig. 2. Effect of the U_4O_9 Content on the Onset Temperature. (Ref. 9)