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# PROCESSING OF HIGH-FIRED URANIUM DIOXIDE FUELS BY A REDUCTION–MERCURY EXTRACTION–OXIDATION PROCESS

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**OAK RIDGE NATIONAL LABORATORY**

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## ABSTRACT

A preliminary flowsheet for the purification of uranium dioxide fuels by a magnesium reduction—mercury extraction—steam oxidation process is proposed. Laboratory-scale scouting experiments indicated the process to be feasible. Data evaluation indicated 100% reduction of uranium dioxide by magnesium although this figure was not demonstrated, chiefly because of poor choice of materials and design of equipment. Steam oxidation of uranium tetramercuride produced an oxide with an O/U ratio of 2.43. This ratio was decreased to 2.09 by heating the oxide in a hydrogen atmosphere at 900°C for 1 hr. The final product had a surface area of 3.5 $m^2/g$, and 18% of the particles were $<1$ μ dia. A pellet of the oxide sintered at 1750°C had a density of 9.76 g/cc, 89% of theoretical. Decontamination factors demonstrated for ruthenium, cesium, and samarium, when present originally in amounts equivalent to 30,000 Mwd/ton fuel burnup and 60 days' decay, were $>10^3$, 220, and 75, respectively.

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## 1.0 INTRODUCTION

The purpose of this investigation was to determine the feasibility of adapting the Hermex process[1-3] to the purification of high-fired uranium dioxide fuels. One of the needs of a fast uranium-fueled power reactor is for a short fuel processing cycle to minimize the inventory of U-235. Pyrometallurgical processing methods have been developed for metal fuels; however, since the use of less expensive, more stable $UO_2$ fuels has received increased emphasis, a need has arisen for a short-cycle processing method for oxide fuels. In this feasibility study, experiments on the reduction of uranium dioxide, extraction of the uranium by mercury, and reoxidation of the uranium to the dioxide, were conducted on a laboratory scale. Results are only semiquantitative. The feasibility of each step of the process was indicated, but considerable study remains to be done, particularly on the extraction and $UHg_4$ oxidation steps.

Reduction of uranium dioxide by magnesium to uranium powder has been reported.[4] In order to obtain a nonpyrophoric material, a flux, anhydrous magnesium chloride, was added to make the magnesia matrix product fluid and to permit the coalescence of fine uranium particles. Reduction of uranium dioxide by magnesium amalgam, even with boiling amalgam, gave low yields. Part of this difficulty is believed due to the resistance of the oxide to wetting by the amalgam. Bellamy and Buddery[4] found that a temperature of about 730°C was required for magnesium reduction. Pressurization of the amalgam reduction reactor to achieve this temperature might increase the yield of amalgam reduction of $UO_2$ to a practicable level.

Extraction of uranium from a mixture of the products of magnesium reduction of uranium dioxide is an extension of the Hermex[1,2] process for purification of uranium metal fuels by mercury dissolution and recrystallization. Volatile fission products would be expected to appear in the off-gas while metals whose oxides are unreduced would remain with the slag. The bulk of the more noble fission products would also remain with the slag as a consequence of their limited solubility in mercury.[3]

Steam oxidation of uranium metal is well known. With proper precautions, such as exclusion of air, the oxidation can be stopped at the dioxide.[5] Because of the extreme reactivity of $UHg_4$ to oxygen, extended study of variables controlling rates and products of oxidation will be required.

The authors are indebted to G. R. Wilson, W. R. Laing, and G. W. Leddicotte and staffs of the ORNL Analytical Chemistry Division for analyses performed. The authors also acknowledge the technical assistance of E. R. Johns in the experimental studies.

## 2.0 PROCESS FLOWSHEET

The flowsheet for processing of uranium dioxide fuels by reduction, mercury extraction, and oxidation (Fig. 2.1) is based entirely on the feasibility experiments described in Sect. 3.0. After a more thorough study has been made, considerable modification of the process may be necessary.

The reduction mixture is irradiated uranium dioxide intimately mixed with magnesium metal (115% of the theoretical requirements for complete reduction) and a flux of anhydrous magnesium chloride amounting to 10% of the $UO_2$ weight. Heating the mixture in a sealed bomb for 2 hr at 1000°C produces a nonpyrophoric uranium powder in the form of 0.5- to 50-μ-dia spheres.

After the mixture has cooled, the uranium is dissolved in boiling mercury along with excess magnesium and to a limited extent the noble metals,[3] notably ruthenium. Fission products such as xenon and cesium volatilize from the mixture during the uranium dissolution. Filtration at 356°C removes a dross containing magnesium oxide and chloride, compounds of unreduced fission products such as strontium and the rare earths, iodine as the iodide,[7] and the majority of the noble metals as insoluble mercurides. As the filtrate cools, the mercurides crystallize and are removed from the excess mercury by filtration at 25°C. The thixotropic uranium quasi-amalgam obtained contains magnesium and noble metal impurities. These impurities are decreased to a low value by redissolving the quasi-amalgam in boiling mercury, recrystallizing the $UHg_4$ at 25°C, and filtering again.

Noble metals are not expected to be affected by steam oxidation of the uranium mercuride at 200°C. Instead, they would be extracted into additional boiling mercury and separated from the oxide product by filtration at 356°C. This mercury filtrate and that from the two 25°C recrystallization steps would be purified and recycled.

The oxide product would contain significant amounts of mercury, which is removed by heating in a retort at 900°C for 2 hr. If the oxygen/uranium ratio is too high, it could be lowered by introducing hydrogen during the retorting. The final product is expected to be quite free of impurities.

Purification of mercury prior to recycle involves oxidation of contained material such as magnesium and filtration at 25°C or distillation.

## 3.0 EXPERIMENTAL RESULTS

Three experiments were performed in which 1750°C-fired uranium dioxide powder and pellets were reduced by magnesium to uranium and the reduction products were leached with boiling mercury. The reductions were 58, 78, and 93% complete as measured by the amount of uranium extracted into the mercury phase. While the low percentage in the first reduction was due to volatilization

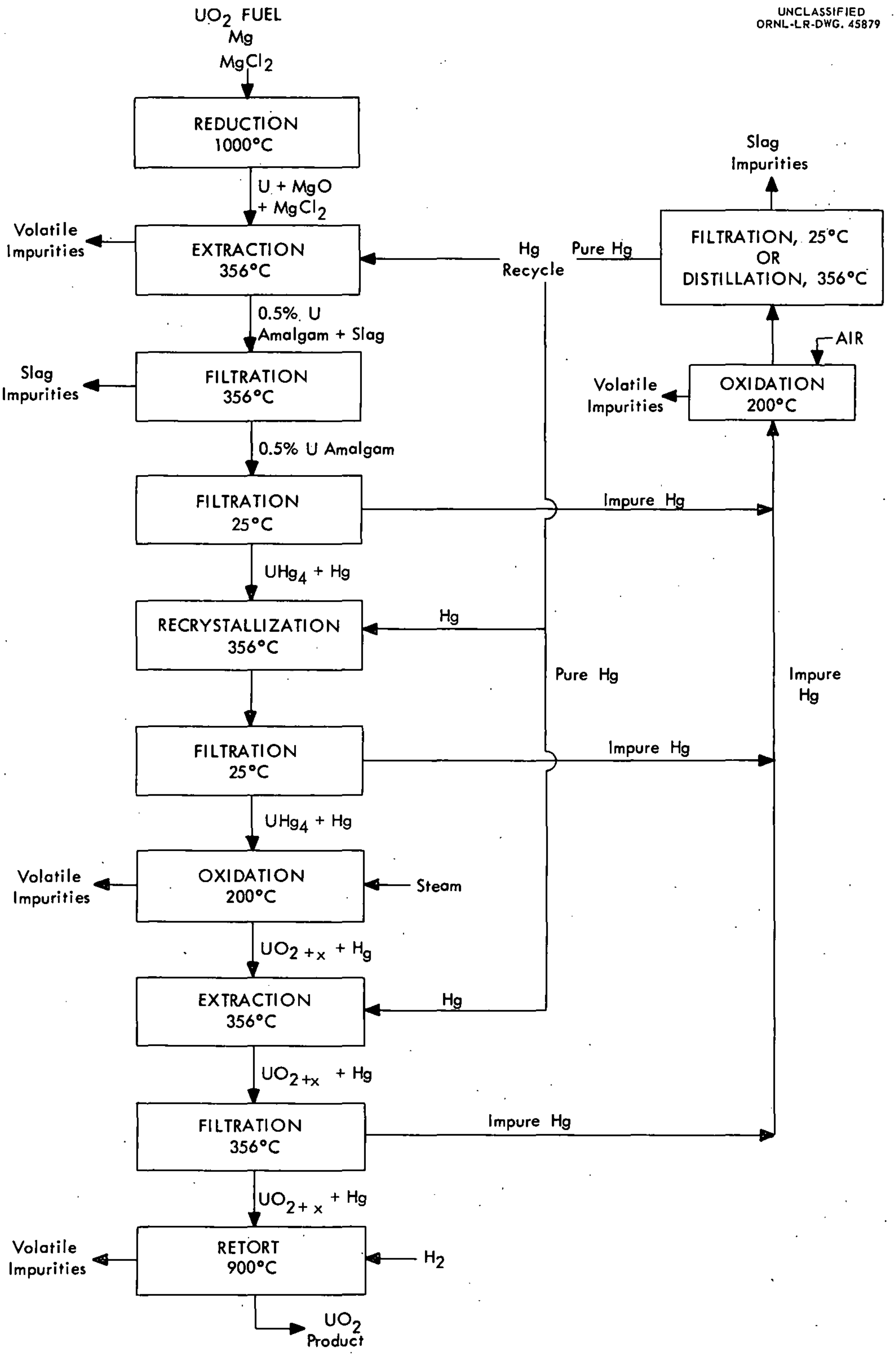

Fig. 2.1. Schematic flowsheet for purifying uranium dioxide fuels by a reduction–mercury extraction–oxidation process.

of magnesium from a leaky reactor, the incompleteness of the second and third was thought to be due to blinding of some of the uranium to the mercury by a coating of magnesium chloride. A similar phenomenon was observed by Schneider[6] for cadmium extraction of the reduction product at temperatures below the melting point of magnesium chloride.

The amalgam extracts, containing $UHg_4$, from one reduction-extraction experiment were combined and treated with steam at 200°C. The $UHg_4$ was converted to uranium oxide with an O/U ratio of 2.43. When heated for 1 hr at 900°C in a hydrogen atmosphere, the O/U ratio was lowered to 2.09, and $>99\%$ of the residual mercury was volatilized from it. The oxide product had a surface area of 3.5 $m^2/g$ and 18% of its particles were $<1$ μ dia. A pellet pressed from this oxide at 15,000 psi and sintered at 1750°C had a density of 9.76 $g/cm^3$, 89% of theoretical.

In the reduction experiment carried through steam oxidation, the $UO_2$ was spiked with ruthenium, cesium chloride, and samarium oxide to the level expected for 30,000 Mwd/ton irradiation and 60 days' decay. Decontamination factors measured from initial to final oxides were $>10^3$ for ruthenium, 220 for cesium, and 75 for samarium.

In attempts to reduce active (6 $m^2/g$) $UO_2$ with magnesium and calcium amalgams at 356°C, reduction yields were less than 1% in 1- and 5-day agitation periods.

## 3.1 Reduction of Uranium Dioxide

By Magnesium. The extent of bomb reduction by magnesium was determined by the amount of uranium extracted from the reaction mixture with boiling mercury. In no case was 100% of the uranium extracted, but, on the basis of unoxidized magnesium that appeared in the mercury extracts, reduction was probably complete.

In Expt. I, only 56.6% of the uranium was extracted from the reduction reaction mixture. No magnesium was found in the mercury extracts while that found with the extraction residue was equivalent to only 58% reduction plus the amount originally added as a flux (Table 3.1). Apparently the bomb had a poor seal and magnesium was lost by volatilization at the reduction temperature.

In Expt. II, 93% of the uranium was extracted by boiling mercury, indicating 93% reduction. On the basis of magnesium that appeared in the mercury extract and that which remained with the residue, reduction was complete (Table 3.1). That extraction was incomplete is indicated by the fact that the amount of mercury used in the first of two extractions was sufficient to dissolve all the uranium if present as the metal. Only 50% was extracted, but an additional 43% was found in the filtrate of the second extraction.

Table 3.1 Magnesium Reduction and Mercury Extraction, Experiments I and II

Reduction temperature: 1000°C for 2 hr
Extraction and filtration temperature: 356°C

| Experiment | | Materials Added and Analyses | | |
|---|---|---|---|---|
| | | U, g | Mg, g | Hg, ml |
| I Reduction bomb initially charged with 27.0 g of 1750°C-fired $UO_2$ pellets, 6.00 g of Mg, and 2.70 g of $MgCl_2$ | | | | |
| Initial bomb charge | | 23.8 | 6.69 | -- |
| Mercury extraction | 1 | -- | -- | 100 |
| | 2 | -- | -- | 35 |
| | 3 | -- | -- | 90 |
| | 4 | -- | -- | 70 |
| Extraction residue | | 10.35 | 3.52 | 5 |
| Extraction filtrate | 1 | 5.32 | 0 | 35 |
| | 2 | 7.85 | 0 | 90 |
| | 3 | 0.37 | 0 | 70 |
| | 4 | 0.27 | 0 | 90 |
| II Reduction bomb initially charged with 18.9 g of 1750°C-fired $UO_2$ powder, 4.18 g of Mg, and 1.97 g of $MgCl_2$ | | | | |
| Initial bomb charge | | 16.6 | 4.69 | -- |
| Mercury extraction | 1 | -- | -- | 110 |
| | 2 | -- | -- | 100 |
| Extraction residue | | 1.19 | 3.71 | 2 |
| Extraction filtrate | 1 | 8.32 | 0.75 | 78 |
| | 2 | 6.54 | 0.16 | 121 |

The final magnesium reduction, Expt. III, was spiked with ruthenium, cesium, and samarium in amounts equivalent to 30,000 Mwd/ton burnup and 60 days' decay. The product of reduction and extraction was steam-oxidized (Sect. 3.2) and the degree of decontamination determined (Sect. 3.3). Reduction based on uranium extracted was 78.3% but this low value is also believed due to incomplete extraction. Based on magnesium present in the extracts and that left in the residue, reduction was 100% (Table 3.2). During the dissolution of the extraction residue, a quasi-amalgam-like material was noted. This material was not analyzed but its appearance was considerably different from that of products in other studies. It is believed to have been uranium metal with a protective magnesium chloride coating[7] which hindered extraction by mercury.

Table 3.2 Magnesium Reduction—Mercury Extraction—Steam Oxidation, Experiment III

Reduction temperature: 1000°C for 2 hr
Extraction and filtration temperature: 356°C
Oxidation by steam at 200°C

| | Materials Added and Analyses | | | | | |
|---|---|---|---|---|---|---|
| Experiment | U, g | Mg, g | Ru, mg | Cs, mg | Sm, mg | Hg, ml |
| III Reduction bomb initially charged with 99.0 g of 1750°C-fired $UO_2$ powder, 20.7 g of Mg, 10.0 g of $MgCl_2$, 290 mg of Ru, 475 mg of CsCl, and 60 mg of $Sm_2O_3$ | | | | | | |
| Initial bomb charge | 87.3 | 23.22 | 290 | 375 | 52 | -- |
| Mercury extraction 1 | -- | -- | -- | -- | -- | 300 |
| 2 | -- | -- | -- | -- | -- | 300 |
| 3 | -- | -- | -- | -- | -- | 200 |
| Added prior to oxidation | -- | -- | -- | -- | -- | 200 |
| Extraction residue | 18.9 | 20.75 | 112 | 295 | 44 | 40 |
| Extraction filtrate[a] 1 | 0.05 | 0.01 | 16 | 0 | 0 | 120 |
| 2 | 0.13 | 0.01 | 45 | 0 | 0.02 | 330 |
| 3 | 0.10 | 0.01 | 34 | 0 | 0.12 | 250 |
| Filtrate of oxide extraction | 6.05 | 0 | 60 | 0 | 0.16 | 225 |
| Miscellaneous[b] | 12.8 | 2.24 | 12 | 2.2 | 0.3 | 40 |
| Oxide product | 49.5[c] | 0.17 | 0.2 | 1.0 | 0.4 | 0.11 |

[a] Extracts cooled to 25°C prior to filtering.

[b] Includes materials from (1) the plugged filter and oxide skimmed from the surface of the initial reduction-extraction amalgam and (2) residue remaining in the oxidation vessel after material transfer.

[c] 56.0 g of $UO_2$.

During the filtration of the first extract of Expt. III at 356°C, mercurides crystallized in the cooler stopcock and ball-joint system leading to the cold filter and formed a plug. Before the filtration could be completed, the equipment had to be disassembled and the plug removed. An oxide that formed on the surface of the filtrate during the time the equipment was disassembled was skimmed off, combined with the material from the plug, and analyzed as "miscellaneous" along with the residue from steam oxidation (Sect. 3.2).

By Magnesium and Calcium Amalgam. Boiling magnesium amalgam (approximately 1 g of magnesium in 4 ml of mercury) and boiling calcium amalgam (approximately 1 g of calcium in 11 ml of mercury) resulted in <1% reduction of active uranium dioxide (6 $m^2$ surface per gram $UO_2$) in 24 hr and 5 days, respectively. The extent of reduction was determined by filtering the reaction mixture at 356°C, re-extracting the mixture with an equivalent amount of boiling mercury, and analyzing the filtrates and residue. Magnesium and calcium were in excess of the amounts required for complete reduction of the $UO_2$ by factors of 13 and 2, respectively.

## 3.2 Steam Oxidation of Uranium Tetramercuride

Steam oxidation of uranium tetramercuride at 200°C followed by retorting at 900°C for 1 hr under 1 atm of hydrogen produced an oxide with an O/U ratio of 2.09. The surface area of the powder was 3.5 $m^2/g$ and 18% of the particles were 1 μ dia. A sample of the oxide pressed into a pellet at 15,000 $lb/in.^2$ was sintered at 1750°C to a density of 9.76 g/cc, 89% of theoretical.

After filtration at 25°C of the mercury extracts from Expt. III, described in Sect. 3.1, 200 ml of mercury was added to the combined mercurides and the temperature raised to 200°C. Steam was passed through the mixture for 1 hr, after which time the temperature was raised to 356°C and the solution filtered. The oxide, which had an O/U ratio of 2.43, was transferred to a nickel boat, placed in a combustion tube, and heated to 900°C under argon flow. Hydrogen was introduced at this temperature and was maintained for 1 hr after the argon flow had been discontinued. The oxide was cooled under hydrogen.

Each gram of the oxide product contained 26 mg of mercury and 3 mg of magnesium (Table 3.2). The mercury could have been completely removed by longer heating in the retort. Magnesium may have coprecipitated with the $UHg_4$ and a recrystallization step might be necessary for its complete removal. Alternatively, its source could have been the oxide which formed on the initial amalgam surface, as described above, which was not completely removed. If so, it would have remained with the $UHg_4$ when the excess mercury was removed by filtration at 25°C.

The mercury filtrate obtained after oxidation contained a significant amount of uranium, indicating that oxidation was incomplete. It is believed that dissolution of some of the mercuride in the mercury at 200°C caused some of the uranium to pass through the filter before and during oxidation. After steam oxidation, mercury was observed above the vessel stopcock and below the filter before the excess mercury was removed by filtration. The small amount of residue remaining in the oxidation vessel after material transfer was dissolved and added to the miscellaneous solution (Sect. 3.1).

### 3.3 Decontamination of Uranium Dioxide by the Magnesium Reduction—Mercury Extraction—Steam Oxidation Process

Ruthenium, cesium, and samarium were added to uranium dioxide in amounts equivalent to a 30,000 Mwd/ton fuel burnup and a 60 days' decay. When carried through the reduction-extraction-oxidation process, decontamination factors of $>10^3$, 220, and 75 were demonstrated for the three impurities, respectively. Greater decontamination is expected with the use of larger samples, better designed equipment, and the inclusion of a mercuride recrystallization step.

The ruthenium, cesium, and samarium were added as the metal, the chloride, and the oxide, respectively, to the original reaction mixture in Expt. III (Sect. 3.1). These amounts are equivalent to contamination of 2.93 mg of ruthenium, 3.79 mg of cesium, and 0.53 mg of samarium for each 1 g of uranium dioxide. Distribution of contaminants during the processing is shown in Table 3.2.

The behavior of ruthenium was essentially as expected on the basis of its solubility in mercury.[3] It was unaffected by steam during the oxidation step since that present followed the mercury removed from the oxide by filtration at 356°C. The final oxide product contained 2.8 μg of ruthenium per gram of $UO_2$, which represented a decontamination factor of $>10^3$. Reextraction of the oxide with mercury would have further lowered the contamination.

At the reduction temperature cesium was expected to be reduced to the metal by the magnesium and to follow the mercury extracts and/or be volatilized during extraction. In reality, the bulk of the cesium was found in the reduction-extraction residue and none in the mercury filtrates. The oxide product contained 17.2 μg of cesium per gram of $UO_2$, a decontamination factor of 220. The decontamination factor would have been greater if the $UHg_4$ had been recrystallized before the oxidation step. Approximately 20% of the cesium was unaccounted for in a material balance. It is thought that magnesium had reduced this material and that it was lost by volatilization during the extraction step. Reduction of cesium may have been greater than indicated. It may have volatilized from the extraction mixture, condensed on a cooler part of the extractor, and then dissolved along with the residue.

Samarium was not expected to be affected by the procedure in the purification process. However, the product oxide was found to contain 7.1 μg of samarium per gram $UO_2$, a decontamination factor of 75. Either some reduction had occurred, or the amalgam peptized the oxide, enabling the above quantity to pass the filter.

## 4.0 EQUIPMENT AND PROCEDURE

The stainless steel reduction bomb was damaged considerably at the reaction temperature of 1000°C. For future experiments, nickel or some other heat-resisting material should be used for bomb construction. During the filtration of a mercury extract at 356°C, mercurides crystallized from solution in the cool portion of the glass transfer line. To overcome this problem, stainless steel should be used for the extraction vessel and the transfer line to the cold filter and a micrometallic filter should be used for filtration at 356°C. Resistance heating may then be used to maintain the temperature required to keep the mercurides in solution during materials transfer.

### 4.1 Equipment

Reductions were performed in 1.5-in.-dia 347 stainless steel bombs fitted with threaded covers. The charge, when fully loaded, was within 1/8 in. of the top. Because of the elevated temperature of the reduction, the threads were galled, making necessary a new bomb for each reduction. A muffle furnace supplied the heat.

The glass equipment was an extractor—hot filter unit, a cold filter, and a mercury collector. The extractor—hot filter and the cold filter were constructed from 500-ml three-necked flasks. The bottom of each was equipped with a fritted glass filter (40 μ effective pore diameter) and a stopcock. Both were fitted with an argon inlet and off-gas line for maintaining an inert atmosphere and a thermowell. The extractor—hot filter was also equipped with a reflux condenser. Glas-Col heaters were the heat source, and the desired temperature was maintained by thermocouples and a Wheelco controller. The mercury collector was a 500-ml vacuum flask. Ball-joint systems connected the extractor—hot filter to the cold filter and the cold filter to the mercury collector.

Amalgam reduction studies were performed in the extractor—hot filter equipped with a mechanical stirrer. The amalgam oxidation studies were conducted in the cold filter after the extractor—hot filter had been replaced with a steam line.

### 4.2 Procedure

A weighed quantity of 1750°C-fired uranium dioxide, magnesium metal (approximately 115% of the amount necessary for complete reduction), and anhydrous magnesium chloride (10 wt % of the $UO_2$) were intimately mixed and placed in the reduction bomb. The sealed bomb was placed in a muffle furnace and raised to a temperature of 1000°C. After 2 hr, the temperature was lowered to 25°C and the reaction mixture transferred to the extractor—hot filter under an argon atmosphere. Mercury was added and the temperature raised to boiling. After several hours the boiling mixture was filtered with the aid of argon pressure. The filtrate was allowed to cool to 25°C and then refiltered in order to separate the precipitated uranium mercuride from the bulk of the mercury. Additional extractions were made as thought necessary at the time.

The extent of reduction was determined by the amount of uranium extracted from the reaction mixture. Therefore reported reduction results may not truly reflect the completeness of reduction.

There was considerable difficulty in filtering the mercury extracts at 356°C. When the extract passed through the cool stopcock, mercurides crystallized and formed a plug which was difficult or impossible to dislodge.

Oxidation was accomplished by passing steam through a suspension of uranium mercuride in mercury at a temperature of 200°C for 1 hr. Oxides formed floated to the surface of the mercury and steam remained in the vapor state until condensed in the off-gas line. After the oxidation, the temperature of the mercury was raised to boiling and the mixture filtered by a combination of argon pressure and vacuum.

The oxides were transferred to a nickel boat and heated in a combustion tube to a temperature of 900°C in an atmosphere of hydrogen and argon. Argon flow was discontinued and the temperature maintained for 1 hr. During this period, mercury was expected to be removed and higher oxides of uranium reduced to the dioxide.

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