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# DECONTAMINATION OF THORIUM-URANIUM ALLOYS BY CONSUMABLE ARC-MELTING

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# DECONTAMINATION OF THORIUM-URANIUM ALLOYS BY CONSUMABLE ARC-MELTING

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## ABSTRACT

A study of the decontamination of thorium-uranium alloy by the consumable-electrode arc-melting process has been carried out. Cesium and strontium removals of up to 75% were obtained, whereas cerium and rare earth removals were less than 20%. Metal loss increases with increasing power to the arc.

The method is not suitable for a decontamination process but could be utilized as a step in a low-decontamination, reprocessing method.

## I. INTRODUCTION

Since thorium is considered as a fissile material for reactors and appears to have an assured place in the future market, industry will eventually be faced with the necessity of decontaminating and reprocessing spent thorium fuels. The procedure for accomplishing this should be established well in advance of its practical use. Of the many methods of decontamination which have been suggested and are being investigated, arc melting is one of the interesting low-decontamination methods. In this procedure, the volatile fission products are evaporated from the molten thorium in an electric arc.

The vapor pressures of representative elements at 2000°K are given in Table I. It can be seen from the data that cesium, tellurium, and strontium would be easy to remove by arc melting, whereas uranium, zirconium, thorium, protactinium, and ruthenium would be difficult to remove by this method.

TABLE I

VAPOR PRESSURE
OF REPRESENTATIVE ELEMENTS AT 2000 ° K[1]

| Element | Vapor Pressure (atm) |
|---|---|
| Cesium | >1 |
| Tellurium | >1 |
| Strontium | >1 |
| Cerium | $4 \times 10^{-2}$ |
| Lanthanum | $10^{-3}$ |
| Uranium | $<10^{-6}$ |
| Zirconium | $<10^{-6}$ |
| Thorium | $<10^{-6}$ |
| Protactinium | $<10^{-6}$ |
| Ruthenium | $<10^{-6}$ |

Murbach and Buyers substantiated these conclusions in a preceding study.[2] Their work was done under helium at a pressure of 1/2 atm, with a nonconsumable electrode. It seemed feasible to continue the arc-melting studies with a consumable electrode and under high-vacuum conditions. It seemed advisable also to determine the amount of uranium or thorium which would be lost by evaporation under high-vacuum operating conditions. The following three sets of experiments were carried out:

1) An irradiated Th-5.4 w/o U electrode was melted in a consumable arc under helium at a pressure of 1/2 atm.

2) An unirradiated Th-10 w/o U electrode was melted in a consumable arc in vacuo at various power conditions.

3) An irradiated Th-5.4 w/o U electrode was melted in a consumable arc in vacuo.

## II. EXPERIMENTAL METHOD

### A. APPARATUS

The consumable-electrode arc-melting apparatus is shown in the Figure. The vacuum is obtained with a Welch fore pump and an MCF-60 oil diffusion pump. The body of the furnace is copper with water-cooling coils soldered around it. The bottom of the furnace is a water-cooled copper hearth which seals to the furnace with an O-ring. The copper electrode holder is water cooled. The holder enters the furnace through a hole in the top; it is held there by an expansion bellows which is sealed to the top with Apiezon wax. The electrode holder then passes through the bellows; high-vacuum grease is utilized as a lubricant and seal.

A window was provided in the furnace for observation. Pressure gages were placed in the system and a filter was placed on the exhaust of the fore pump to prevent contaminants from reaching the room atmosphere. A 300-amp P & H dc arc welder, Model 302 was used as a power supply. It is capable of giving currents ranging from zero to well over 300 amp. The hearth was made negative with respect to the electrode and a voltmeter and ammeter were placed in the line for controlling parameters.

### B. PROCEDURE

The experimental procedure is summarized as follows:

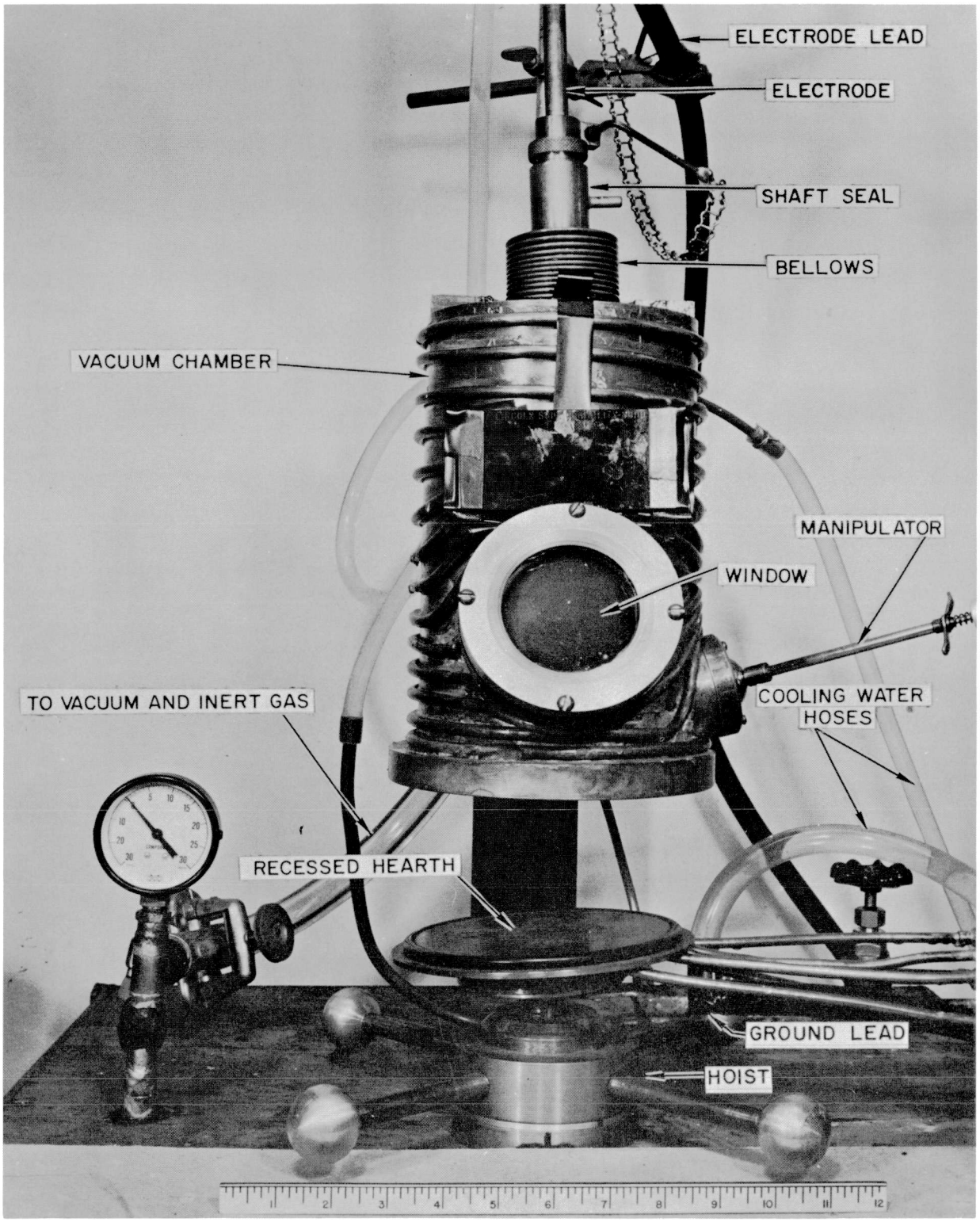
1) An electrode of Th-U alloy was placed in the holder and the set screws were adjusted to hold the electrode in place.

2) The hearth was raised with the screw jack so that a seal was made with the furnace body.

3) The fore pump was started and the diffusion pump was turned on when the pressure dropped below 100 microns.

4) The furnace cooling water was turned on.

5) The power supply leads to the hearth and electrode holder were connected.




Consumable-Electrode Arc-Melting Apparatus

6) For the first set of experiments, the system was evacuated, flushed with helium, and adjusted to 1/2 atm of helium. For the second and third sets of experiments, the system was merely evacuated.

7) The power supply was turned on.

8) An arc was struck by depressing the electrode holder so that the electrode touched the hearth, and the arc was adjusted to the proper length. It was found originally that arcing took place in vacuo to all parts of the electrode holder from all parts of the furnace. It had the appearance of a glow discharge. The hearth was further insulated from the body of the furnace by using a silicone gasket over the O-ring. In this state, only the hearth and electrode holder were charged and arcing proceeded normally.

9) The voltage and current were adjusted to the proper values and recorded.

10) When a sufficient quantity of the electrode had melted and dropped to the hearth, the power supply was turned off.

11) After cooling, the hearth was lowered and the sample was removed from the furnace.

12) Samples of each melt were taken for analysis. The samples were dissolved in nitric acid with a grain of fluoride salt as a catalyst. The solution was then analyzed for fission products by standard methods. Samples of unmelted material were also analyzed for comparison.

## III. RESULTS

The first experiment was the consumable melting of irradiated Th-5.4 w/o U under helium at 1/2 atm. The results ot the analyses of the melt for fission products are given in Table II.

TABLE II

CONSUMABLE-ELECTRODE, ARC-MELTED, IRRADIATED Th-5.4 w/o U IN HELIUM AT A PRESSURE OF 1/2 Atm

| Run | Amount Melted (gm) | Amount Removed (%) | | | | |
|---|---|---|---|---|---|---|
| | | Sr | Cs | Ce | Re | Ru |
| 1* | 16.2 | 64 | 40 | 9 | 6 | 0 |
| 2† | 21.4 | 75 | 73 | 17 | 16 | - |

* The melt was spread on the hearth.
† The melt was compacted as a button.

The second experiment was the consumable melting of unirradiated Th-10 w/o U in vacuo at various power outputs. The electrode weights were determined before and after melting. The results are given in Table III.

TABLE III

CONSUMABLE-ELECTRODE, ARC-MELTED, UNIRRADIATED Th-10 w/o U IN VACUO AT VARIOUS POWER OUTPUTS

| Sample | Force (v dc) | | Current (amp) | Pressure ($mmHg \times 10^{-4}$) | Amount (gm) | Amount Lost (gm) | Amount Lost (w/o) |
|---|---|---|---|---|---|---|---|
| | Arc | No Arc | | | | | |
| 1 | 20 | 72 | 110 | 0.19 | 32.9974 | 0.0259 | 0.078 |
| 4 | 20 | 72 | 150 | 1.0 | 45.0075 | 0.0946 | 0.21 |
| 2 | 20 | 72 | 200 | 3.0 | 50.6228 | 0.1733 | 0.34 |
| 5 | 20 | 72 | 255 | 2.0 | 37.0241 | 0.0902 | 0.24 |
| 3 | 20 | 72 | 300+ | 0.93 | 46.6099 | 0.3295 | 0.71 |

The third experiment was the consumable melting of irradiated Th-5.4 w/o U in vacuo at various power levels. The melted and unmelted parts were analyzed by the analytical lab for cesium, strontium, and total activity. The results are given in Table IV.

TABLE IV

CONSUMABLE-ELECTRODE, ARC-MELTED, IRRADIATED TH-5.4 w/o U IN VACUO AT VARIOUS POWER OUTPUTS

| Sample | Current (amp) | Amount Removed (%) | | |
|---|---|---|---|---|
| | | Strontium | Cesium | Total Activity |
| 6 | 120 | 51.0 | 61.3 | 16.3 |
| 7 | 200 | 71.7 | 73.5 | 29.9 |
| 8 | 300+ | 55.8 | 48.2 | 21.2 |

## IV. DISCUSSION

Table II indicates that the losses of the fission products follow the vapor pressure of the elements in the case of the consumable electrode as found during previous button experiments.[2] The decontamination as shown in Tables II and IV is not as good with a consumable electrode as arc melting a button. The reason for this is probably the length of time which the alloy is molten in each case. The dropping, molten metal is more favorable to the loss of fission products by evaporation, due to its larger surface area; however, the time element favors the button arc melting. Since the duration of the molten state is the determining factor, greater decontamination was obtained in the button experiments previously reported.

It can also be noted from Table II that a consumable electrode which is spread out along the hearth is not decontaminated as much as one which is compacted as a button. This is because the electrode which is spread out as it is melted loses heat to the water-cooled hearth faster than one which is melted into a button. This result corresponds with that which was expected.

The results recorded in Table III show the weight loss of alloy as a function of power level (amperes in the arc). It is assumed that the temperature of the melt is proportional to the power level since it was very difficult to take temperature measurements. It was not known if there would be an effect or what effect there might be. It can be seen from the data that there is an effect which can become appreciable at high power levels. The results for sample number five are out of line. It is thought that this is due to fluctuations in the arc during melting. The arc sputtered and on one occasion went out, as also happened with Sample 8. As expected, the loss of weight increases with an increase in current or power level.

The third and last experiment was a decontamination study under high vacuum and various power levels. Comparison of Tables I and III indicate that the effect of vacuum is small in comparison to the effect of current. In fact, the effect of current so masks everything that it is not possible to decide what the effect of vacuum is. It is reasonable to assume that a greater decontamination would result in high vacuum than under an atmosphere since there is no resistance to

the evaporation of fission products in vacuum and that the effect would be small since the partial pressure of the fission products is the same in each case.

The effect of power is that as the power is increased, the decontamination is increased. Sample number eight is inconsistent, probably due to fluctuations in the arc. Since an increase in power increases the decontamination and the thorium or uranium loss, it does not seem feasible to use this as a method of increasing decontamination. Decontamination could conceivably be increased by eliminating the water-cooled hearth and using a preheated hearth. This would keep the thorium in the molten state for a longer period of time. If this were done, however, another problem would be inevitable. Molten thorium will react with most metals and refractories if given sufficient time. The contamination from this reaction might prove to be more serious than the fission products.

## V. CONCLUSIONS

It has been shown that the consumable-electrode arc-melting process as a means of decontaminating thorium-uranium alloys is in general inferior to the nonconsumable arc-melting process. The method is applicable only to the removal of the volatile fission products. When the temperature is taken high enough to remove other impurities, losses of vital metal become appreciable. It could be used as one step in a decontamination process since it lends itself to ease of operation. A possible use of the process could be in the refabrication of metal partially decontaminated by another process.

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2. E. W. Murbach and A. G. Buyers, "Decontamination of Thorium-Uranium Alloy by Nonconsumable Electrode Arc-Melting," NAA-SR-1989, September 1, 1957.