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FMD Classified H-331-13 Thorium

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PRODUCTION OF THORIUM METAL: AMES PROCESS FOR REDUCING $ThCl_4$ WITH MAGNESIUM

SYMBOL: PP:JPT

During my visit to Ames Iowa State College on March 4 and 5 to discuss principally monazite processing, I also reviewed with Dr. H. Wilhelm and Dr. D. Peterson of Ames Iowa State College their experimental work on $ThCl_4$ preparation and its reduction with magnesium to thorium metal.

Summary:

The work to date has been only on a laboratory scale. Approximately 20 to 25 lbs. of $ThCl_4$ have been produced by chlorinating ThO_2 and C in batches of approximately 250 to 300 grams with 95% to 99% yields. The preparation of high purity $ThCl_4$, free from ThO_2 and carbide contaminants, has been a problem.

About 10 to 15 lbs. of sponge Th, containing 2% to 50% magnesium, has been made in a 2½" i.d. x 12" steel bomb lined with a pre-sintered MgO and CaF_2 and filled with argon. The purity and quality of the Th metal product has not been established. Scoping experiments indicate, however, that purification of the $ThCl_4$ by fractional distillation may be necessary to increase the $ThCl_4$ density and to reduce the impurities of the Th metal. Ames personnel believe that a Th sponge, containing 2% to 3% magnesium and suitable for vacuum casting, could be produced by this method with yields of greater than 95%.

At present, work on this project has been discontinued, but Drs. Wilhelm and Peterson both expressed a keen interest in continuing this development on a larger scale.

Classification Cancelled
~~Or Changed To~~
By Authority Of DOC
By Ted Davis Date 8-29-85

Discussion:

A. Preparation of $ThCl_4$:

1. Reaction:

$$ThO_2 + 2C + 4Cl_2 \longrightarrow ThCl_4 + 2COCl_2$$

2 hours at 1100°C and an additional 5 hours at 800°C

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2. Equipment:

A porous carbon crucible, containing the ThO_2 and C charge, is located at the closed end of an induction-heated quartz thimble of 4" opening and 24" length. The chlorine is injected at the open end of the thimble, where the $ThCl_4$ product is condensed and collected.

3. Experimental Work:

The charge has ranged from 250 to 300 grams; the total $ThCl_4$ produced has been approximately 20 to 25 pounds. Almost complete reaction occurs, leaving less than 1% of the charge as ash.

Large excesses of chlorine have been used; the quantity required for complete reaction has not yet been determined.

Contamination of $ThCl_4$ is believed to be the result of the mechanical carry-over of the ThO_2 and C. Although the use of a porous carbon crucible has reduced the quantity of these contaminants in the Th metal product, Ames personnel believe that fractional distillation would be necessary to produce a purified $ThCl_4$.

A larger scale study is necessary to determine the temperatures required, the concentrations and quantities of the reactants, and the problems, if any, in separating the $ThCl_4$ from the excess chlorine and the phosgene produced.

I mentioned that Horizons, Inc., under a contract with NYOO Research Division for the study of the electrolytic process for Th production, was also developing a method of $ThCl_4$ and suggested that Ames review this process.

B. Reduction of $ThCl_4$ with Magnesium:

1. Reduction:

$$ThCl_4 + 2Mg \rightarrow 2MgCl_2 + Th$$

Argon atmosphere; 1 hour @ 950°C to 1000°C

2. Equipment:

$2\frac{1}{2}$" i.d. x 12" steel bomb lined with pre-sintered MgO and CaF_2.

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3. Experimental Work:

Approximately 10 to 15 lbs. of sponge Th containing 2% to 50% magnesium has been produced in batches of 150 to 180 grams of Th per bomb. Ames personnel believe that there would be no serious problem in attaining greater than 95% yield in $ThCl_4$ reduction step and in casting the resulting sponge metal by vacuum melting.

The porosity of the liner material has been a problem. The $ThCl_4$ diffuses into the liner, thereby reducing the effective yield. A non-porous metal liner, such as molybdenum, tantalum, or vanadium, appears necessary.

Because of the low heat of reaction, heat must be supplied to reduce $ThCl_4$ efficiently.

The purity of the Th metal produced by this method has not be established. Carbide and metal impurities from the chlorides have been detected in the metal product. The Th metal produced by this method also appears to be harder (possibly because of carbide content) than the Th produced by the reduction of ThF_4.

C. General:

A formal Ames report covering this project has not been issued.

We briefly reviewed the economics of this method. We agreed that the production of $ThCl_4$ may be more expensive than ThF_4 preparation, but the cost for reducing $ThCl_4$ with magnesium could be considerably less than the calcium reduction of ThF_4. This potential savings suggest that the $ThCl_4$ route be studied further, and Drs. Wilhelm and Peterson both expressed a keen interest in continuing development work on a larger scale.

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