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MAGNESIUM REDUCTION PROCESS FOR PRODUCTION OF URANIUM

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1 Claim. (Cl. 75—84.1)

The present invention relates to the production of uranium metal in massive and substantially pure form.

Heretofore uranium has been produced on an experimental basis by electrolysis of a fused salt, such as KUF_5. The cost of metal so produced was expensive, amounting to about one thousand dollars per pound. Also, since KUF_5 was produced photochemically under the action of sunlight, the supply of this material was uncertain and constituted a potential bottleneck in the production of uranium metal. Because of these two objections, a better method of obtaining uranium was sought which would be cheaper and would dispense with the need for using KUF_5.

Among the alternate methods of producing uranium suggested by the prior art are several attempts to obtain the metal by reacting a salt thereof with an active metal. For example, in the British patent to Westinghouse No. 230,865 (1925) KUF_5 is reduced by means of magnesium metal, the process being carried out under a vacuum. Obviously such a method does not solve the problem of a substitute for KUF_5, nor is it convenient to operate under a vacuum if a method can be evolved for carrying on the reaction in the open air.

Prior to our invention it was also known that uranium in hard brittle form could be obtained by reducing uranium chloride with calcium in a high vacuum. However, the metal produced in this manner was not of sufficient purity for certain uses of uranium, and metallic calcium is expensive and not readily obtainable in quantity.

It is the object of this invention to provide a new, economical and easy method for producing metallic uranium in massive and substantially pure form.

The object of this invention is obtained by reacting uranium tetrafluoride and magnesium metal and supplying sufficient heat in addition to the heat of reaction to permit the uranium metal to coalesce into a coherent mass. By use of our method, it is not necessary to carry on the reaction under a vacuum, and the employment of KUF_5 and calcium is entirely eliminated.

We have found that magnesium metal will reduce uranium tetrafluoride according to the following equation:

$$UF_4+2Mg=2MgF_2+U \qquad (1)$$

By substituting the atomic weights for the several items in Equation 1, we obtain:

$$314.17+48.6=124.6+238.17 \qquad (2)$$

Dividing Equation 2 by 48.6, we obtain the relative weights of materials required for the reaction:

$$6.45+1=2.56+4.9 \qquad (3)$$

From 3 it is apparent that in a theoretically perfect reaction, 1 gram of metallic magnesium and 6.45 grams of uranium tetrafluoride should yield 4.9 grams of metallic uranium and 2.56 grams of magnesium fluoride.

Upon the assumption that about 25% of the uranium tetrafluoride will form a low melting flux and will remain in the slag, we prefer to use more than the theoretical amount of uranium tetrafluoride required. In practice we have found that a ratio of 7.30 grams of uranium tetrafluoride to 1 gram of magnesium gives satisfactory results.

When dealing with materials in amounts in the neighborhood of several kilograms, we have found that the heat of reaction between the uranium tetrafluoride and the magnesium is insufficient to make the resulting uranium metal fluid enough to separate from the slag and form a pool in the bottom of the crucible used. We therefore heat the reaction crucible before charging it with the material for the reaction. We continue to heat the mixture while the reaction is in progress and for a sufficiently long time thereafter to assure separation of molten uranium from the slag.

In practicing our invention we have found that magnesium vapor formed in the reaction effectively protects the materials from the atmosphere, and that the use of a vacuum or a protective atmosphere may be eliminated. It is to be understood, however, that our method may be performed under a vacuum or in an inert atmosphere, such as argon, if desired.

An electrically heated graphite crucible has been found satisfactory for our purposes. This is heated while empty to approximately 1000° C. to 1200° C. A small charge of magnesium is then introduced, after which small charges of uranium tetrafluoride and magnesium are alternately introduced into the crucible until the total charge has been placed therein. After the charging operation is complete, a cover is placed over the charging hole. The initial effect of charging the crucible is a drop in temperature, since the materials absorb heat until the reaction temperature is attained. Once the reaction between the uranium tetrafluoride and magnesium begins, however, the temperature rises sharply. We continue heating the crucible for about an hour after the reaction has taken place to permit the metal formed in the reaction to coalesce into a pool in the bottom of the crucible. Since the melting point of uranium is about 1125° C., the charge should be heated to at least this temperature, and preferably should be heated higher in order to assure good separation of uranium from slag. However, it is important not to heat the charge to a temperature in excess of 1400° C. when using a graphite crucible, because uranium reacts with carbon to form carbides at temperatures above 1400° C.

Our invention may be illustrated in greater detail by reference to the following specific examples:

Example 1.—A graphite crucible approximately 13" high by 6¾" inside diameter was covered with a graphite top having a charging hole therein about 2" square. The crucible was electrically heated to about 1220° C.,

at which time charging was begun. The following data was recorded:

Time, p. m.	KW	UF_4, gr.	Mg, gr.	Temp., ° C.	Remarks
	5.5			1,050	Empty crucible.
	6.5			1,220	Do.
3:56	4.5		190	about 800	
3:58	4.5	1,390		about 800	No reaction observed.
4:01	4.5		180	about 800	
4:03	4.5	1,310		about 800	Mg burned some, and U sparks observed.
4:08	4.5		150	about 800	
4:08	4.5	1,090		about 800	Quiet or no reaction.
4:10	4.5		130	about 800	Do.
4:10	4.5	950		about 800	Do.
4:12½	4.5		120	about 800	Do.
4:12½	4.5	880		about 800	Do.
4:15	4.5		120	about 800	Do.
4:15	4.5	880		about 800	Do.
4:16	4.5		110	about 800	Do.
4:16	4.5	860		about 800	Do.
4:20	4.5				Mg burned violently.
4:32	4.5				Added 110 gr. UF_4 powder to protect metal.
4:38	6.0				
5:38	7.0				Metal feels sandy.
5:57	7.5			1,232	Surface fumes.
6:30	7.5				Metal still sandy. Added another 110 gr. of UF_4.
7:05	7.5			1,350	Shut down.

In the described experiment a total of 1000 grams of magnesium and 7300 grams of uranium tetrafluoride were used in the charge. An additional 220 grams of uranium tetrafluoride were added for fluxing.

When opened, the crucible was found to contain a regulus of uranium weighing 2070 grams and less than .05 percent of magnesium. This amounted to 42.3 percent of the theoretical yield of 4900 grams of metal.

Example 2.—An experiment was conducted in a manner similar to the first example, except for the addition of an argon atmosphere. A graphite tube in the crucible cover adjacent the charging hole was used for introducing the gas into the crucible. The argon atmosphere was

maintained both while the reaction was taking place and during the cooling period. By this method 6055 grams of uranium tetrafluoride yielded about 1600 grams of metal.

It will be understood that we intend to include variations and modifications of the invention and that the preceding examples are illustrations only and are not to be construed as limitations upon our invention, the scope of which is defined in the appended claim.

We claim:

The method of producing substantially pure massive metallic uranium that comprises the heating of a charge of uranium tetrafluoride and magnesium to cause reaction between them with reduction of uranium fluoride to uranium metal and formation of magnesium fluoride, continuing heating after reaction and maintaining the reacted charge at a temperature above at least 1125° C. for sufficient time for separation of liquid uranium and slag into two layers, said operations being carried out at substantially atmospheric pressure.

References Cited in the file of this patent

UNITED STATES PATENTS

1,415,516	Bridge	May 9, 1922
1,437,984	Marden	Dec. 5, 1922
1,602,542	Marden	Oct. 12, 1926
1,659,209	Marden	Feb. 14, 1928
2,446,780	Newton	Aug. 10, 1948

FOREIGN PATENTS

230,865	Great Britain	Dec. 10, 1925

OTHER REFERENCES

Spedding et al.: USA. E. C. Publication No. CT-2712 date June 26, 1945, Declas. Jan. 4, 1956 pages 1–12, entire publication 67 pages.

Goggin et al.: Industrial and Engineering Chemistry, vol. 18 (Feb. 1926), pp. 114–116.