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MANUFACTURING OF METALLIC URANIUM

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(1) URANIUM OXIDES

(2) COMMINUTION

STARCH + WATER

(3) PREPARING OF A PASTY MIXTURE OF COMMINUTED URANIUM OXIDES AND STARCH

(4) ADDITION OF SILICON METAL POWDER TO THE PASTY MIXTURE IN EXCESS OF THE THEORETICAL QUANTITY

(5) COMPRESSING OF THIS MASS INTO PIECES

(6) HEATING THE COMPRESSED PIECES AT TEMPERATURES OF ABOUT 1700 — 1900° C., THEREBY REDUCING THE URANIUM OXIDES — FORMATION OF A METAL MELT OF URANIUM AND SILICON, PARTLY IN THE FORM OF AN ALLOY AND PARTLY IN THE FORM OF URANIUM SILICIDES

(7) THERMAL TREATMENT AT TEMPERATURES APPROXIMATELY BETWEEN 1100 AND 1400°C. TO CONVERT THE SILICIDES INTO AN ALLOY

(8) COMMINUTION OF THE THUS TREATED MATERIAL TO POWDER

(9) EXTRACTION OF THE SILICON FROM THE ALLOY WITH DILUTE AQUEOUS ALKALINE SOLUTION — SEPARATION OF THE URANIUM METAL POWDER SKELETON FROM THE SOLUTION, WASHING, AND DRYING

(10) METALLIC URANIUM

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MANUFACTURING OF METALLIC URANIUM

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This invention relates to a process for the manufacture of metallic uranium.

The manufacture of metallic uranium is fraught with many difficulties. The reduction of uranium oxides in the electric furnace, using coal as a reducing agent, leads to the formation of undesirable carbides. Direct reduction of the oxides with calcium or calcium hydride has not proved to be practical on a commercial scale. In view of such shortcomings, other methods of producing metallic uranium were sought. Halogen compounds were formed from the oxides first, and from the halogen compounds, uranium was then separated by reduction with sodium and calcium. But these methods, too, are cumbersome and expensive.

The primary object of the present invention is to generally improve the manufacture of metallic uranium. A more specific object is to provide a process which will make it possible to reduce uranium oxides to metal without the roundabout way over the halides. Other objects of this invention will appear from the following description.

In accordance with my present invention, its objects are achieved by comminuting the starting uranium oxides, working the comminuted oxides with a reducing gelatinous preparation into a paste, thoroughly mixing the mass thus obtained with silicon powder, and compressing the mixture into one or more individual pieces. The compressed pieces are heated and subjected to a reducing action and yield a melt bearing uranium and silicon. The silicides in the melt are converted by a thermal aftertreatment into an alloy which will contain the silicon in a form that is extractable by alkaline liquors. The alloy is reduced to a finely divided form, the silicon is extracted by means of alkaline liquors, and the residual skeleton of uranium metal powder is separated from the solution, washed, and dried.

My process as outlined hereinbefore consists of a series of operations which appear epitomized in the flow sheet accompanying the specification. Referring to the flow sheet, the following will explain the various operations and will also serve as a specific example of the process of the invention.

(1) Uranium oxides are the starting material. I prefer to use U_3O_8, but other oxidation levels may be considered as well, provided they are readily available.

(2) Comminution: For instance, U_3O_8 is comminuted in a dry rolling crusher, using rolling wheels of hard manganese steel, equipped with a screen having 10,000 openings per square centimeter (German standard DIN 100). Any other suitable crusher may also be used for the comminution of the uranium oxides, provided no contaminating matter is freed from the mill during operation. Milling is followed by screening.

(3) Preparation of a paste: The comminuted oxides are worked with a reducing gelatinous preparation, for instance, a jelly of potato starch flour, into a paste.

The paste is prepared, for instance, as follows: Of the

weight of the U_3O_8 powder, approximately 2% potato starch flour, such as Knorr potato starch flour, are required. Thus, 6 parts by weight of potato starch flour are needed for 300 parts by weight of U_3O_8.

For each 100 parts by weight of U_3O_8, 50 parts of cold water are needed. The calculated amount of potato flour is gradually stirred into the water which is then heated to boiling. A jelly is formed into which the U_3O_8 powder is mixed. This procedure aims at having each U_3O_8 particle well enveloped by the jelly. I have found that this is necessary if the subsequent reduction is to be performed successfully and without excessive formation of dross.

(4) Addition of silicon powder: Metallic silicon obtained in powder form from the purification is mixed into the U_3O_8-bearing gelatinous mass by kneading and mixing. The preferred proportion of silicon to U_3O_8 is one part of silicon for 1.33 parts of U_3O_8.

(5) The preparation of compressed pieces may be done, for instance, as follows:

(*a*) Drying: The Si and U-bearing jelly obtained by operation (4) is dried at a temperature not exceeding 100° C. For this purpose, the gelatinous mass may be spread in form of a cake, for instance, on a sheet of metal, the bottom of which has previously been sprayed with machine oil to facilitate the release of the dried material. The metal sheet may be filled to two thirds of the height of its raised edge.

(*b*) Coarse granulation and compressing: The dried cake is crushed and then ground, for instance, in a special granulator to coarse granules. The latter are moistened with water, for instance, by spraying. The moistening is to be controlled so that the material will not lump together but will just loosely cohere. The material is then compressed, and the compressed pieces are stored in a dry place.

(6) Heating and reduction of the compressed pieces: This may be done, for instance, as follows:

(*c*) The compressed pieces are placed in a thoria crucible, the inner walls of which have previously been dusted with silicon powder of extreme purity, and are covered thereafter with high melting salts, preferably borax. The crucible is then set into a high frequency furnace.

(*d*) Reduction: The crucible and its contents are heated in the furnace to approximately 1,700 to 1,900° C., preferably 1,800°, for an extended period, preferably two hours. A metallic melt covered with slag is obtained. It consists of uranium and silicon, which are present partly as an alloy, partly in the form of uranium silicides.

(*e*) Pouring: The melt is poured into molds, for instance, plate molds. The molds are turned over upon cooling, and the slag is separated from the solid regulus. Since the slag still holds minor metallic inclusions, it may be treated specially to recover the last beads of metal.

(*f*) Comminution: The metallic regulus which consists, as has already been stated, of an alloy, silicides of uranium, and of silicon, is comminuted, for instance, by crushing in a crusher, and by subsequently granulating in a granulator.

(7) Decomposition of uranium silicides: The uranium silicides are converted into an alloy which will contain the silicon in a form that can be leached by alkali. This is best done in a high-vacuum furnace in the absence of reactive compounds, for instance, in a protective atmosphere of gases of the helium group, such as helium, argon, neon. If no such gas is available, the material may, in an emergency, be treated under a cover of borax. The temperature of treatment may be raised just below the melting point of the alloy, which temperature may be

somewhere between 1,100° and 1,400° C., preferably between 1,200° and 1,300°. The time of treatment is, for instance, three hours. The alloy of uranium and silicon thus obtained is practically free of silicides and contains the silicon in extractable form.

(8) Fine comminution (pulverization): The alloy is reduced to powder so that no foreign metals, such as iron, are introduced (rolling mill with wheels of hard manganese steel or roller mill). The fineness of the powder should correspond, for instance, to the German standard DIN 80 (screen with 6400 openings per square centimeter, 0.075 mm. diameter of the opening).

(9) Extraction of silicon: The finely divided alloy is leached with an alkaline liquor, for instance, a solution of sodium hydroxide, according to the equation

$$Si+2NaOH+H_2O=Na_2SiO_3+2H_2$$

This means that actually one part by weight of NaOH is required to extract approximately 0.35 part Si.

Contrary to nickel, the uranium is extremely sensitive to NaOH (cobalt behaves similarly to uranium in this respect). Whereas with nickel extreme concentrations of NaOH, such as 500 g./l. NaOH, are permissible and extraction may be performed at boiling, such working conditions must not be used with uranium as the latter would go into solution in large amounts together with the silicon. The extraction is therefore performed with diluted liquors containing, for instance, 40 to 70, preferably 50 to 60 g./l. NaOH. The temperatures used are only slightly above room temperature, that is, between 30 and 40° C., preferably 35–36° C. After this treatment, I prefer to run a control analysis to ascertain that all Si was dissolved. Otherwise, leaching must be continued under the conditions mentioned hereinbefore until practically all Si is dissolved.

The uranium metal powder which remains undissolved in the characteristic skeleton form is thoroughly washed in the usual way and then dried.

(10) Final product: The uranium metal powder produced by my process and given thereby a skeleton structure has proved to be an extremely interesting metal, for instance, as regards its unusual magnetic properties and its special suitability as a catalyst for hydrogenation processes.

If the product is to be used as a contact catalyst, it must be stored under ethyl alcohol because it tends to oxidize under the influence of even minute traces of residual alkali.

If the uranium is to be processed further by sintering or casting, it is dried, after washing and filtering, with acetone.

It will be apparent that while I have described my invention in a preferred form, many changes and modifications may be made without departing from the spirit of the invention defined in the appended claims.

Summary of the process

(1) Starting material: Uranium oxides
(2) Comminution of the uranium oxides
(3) Preparation of a paste with a reducing gelatinous preparation
(4) Addition of silicon powder with kneading and mixing
(5) Preparation of compressed pieces:
  (*a*) Drying up to 100° C. and formation of a cake
  (*b*) Coarse granulation, slight moistening with water, and compressing
(6) Heating and reduction:
  (*c*) The compressed pieces are charged into a high frequency furnace and covered with high melting salts
  (*d*) Reduction: Heating to approximately 1,700 to 1,900° C. for an extended period. Formation of a metal melt
  (*e*) Pouring: The hot metal melt consisting of uranium and silicon, partly in the form of silicides, is poured into a mold, and the metallic regulus is separated from the slag
  (*f*) Comminution: The regulus is crushed in a crusher and coarsely granulated in a granulator
(7) Decomposition of the uranium silicides: The silicides are converted by a thermal treatment into a form which is extractable by alkaline liquors
(8) Fine comminution of the mixed alloy of uranium and silicon
(9) Extraction of the silicon with alkaline liquors as alkali silicate, washing, and drying
(10) Final product: Uranium metal powder in form of a metal skeleton

I claim:

1. Process for the manufacture of metallic uranium, comprising comminuting uranium, mixing the comminuted oxides with approximately 2% by weight of starch into a paste, mixing the pasty mixture with silicon powder in excess of the theoretical quantity up to a proportion of approximately 1.33 parts by weight of uranium oxides to one part by weight of silicon, compressing the mixture of uranium oxides, starch and silicon metal powder into pieces, heating the compressed pieces to temperatures of about 1700–1900° C., thereby reducing the uranium oxides and obtaining a melt bearing uranium and silicon partly in the form of an alloy and partly in the form of uranium silicides, subjecting said melt to a thermal treatment at temperatures approximately between 1100 and 1400° C. to convert the silicides into an alloy containing the silicon in a form extractable by dilute aqueous alkaline solutions corresponding to approximately 40–70 g. NaOH per liter, comminuting the material to a finely divided form, extracting the silicon from the material by means of a dilute aqueous alkaline solution of said strength, separating the remaining uranium metal powder skeleton from the solution, washing, and drying.

2. In the process according to claim 1, preparing said paste from water and potato starch flour, incorporating powdered uranium oxides into the jelly by stirring and intimately mixing, and adding pure silicon powder to this mix.

3. In the process according to claim 1, drying the mixture of uranium oxides, paste, and silicon powder, prior to its being compressed, comminuting the dried mixture, moistening with water, and subjecting the moist mixture to said compression.

4. In the process according to claim 1, heating the compressed pieces under a cover of salts which are stable at the temperatures used, separating the silicide-bearing melt from the slag, and pouring the melt into molds.

5. In the process according to claim 4, heating the compressed pieces to approximately 1800° C.

6. In the process according to claim 1, allowing the silicide-bearing melt to solidify, comminuting the solidified mass, and heating the comminuted mass in the absence of reactive substances, in a high-vacuum furnace to approximately 1100 to 1400° C., thereby converting the silicides into an alloy containing the silicon in said extractable form.

7. In the process according to claim 6, heating the comminuted mass in the high-vacuum furnace to approximately 1200 to 1300° C.

References Cited in the file of this patent

UNITED STATES PATENTS

854,018	Becket	May 21, 1907
866,421	Becket	Sept. 17, 1907
874,977	Menges	Dec. 31, 1907
929,578	Fuller	July 27, 1909
982,135	Hansen	Jan. 17, 1911
1,019,394	Weintraub	Mar. 5, 1912