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PROCESS FOR THE PREPARATION OF A URANIUM COMPOUND IN POWDER FORM

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This invention relates to processes for the preparation of powders and in particular, powders of refractory metal compounds, such as uranium carbide, uranium nitride and uranium silicide.

The object of the invention is to make such processes more economical than at present, particularly by dispensing with the grinding step generally necessitated in conventional methods.

According to the invention, a process for the preparation of a refractory metal compound in powder form is provided, which comprises forming a metal-ceramic composition of the compound and of a metal, as hereinafter defined, treating the composition with at least one agent which acts upon the metal to cause disintegration of the composition and separating the desired compound in powder form.

The metal-ceramic composition is preferably formed by sintering the compound and the metal together.

The expression "metal" as used in relation to the material associated with the refractory metal compound in the composition means a metal per se or such metal in alloyed form. It is preferably for the metal to be the same as that in the refractory compound.

In one preferred embodiment of the invention, the agent causes disintegration of the composition by chemical reaction with the metal.

In another preferred embodiment of the invention, the agent causes disintegration of the composition by dissolution of the metal.

In accordance with a preferred feature of the invention, the composition is subjected prior to disintegration to a grain-refining heat treatment.

In carrying out the invention and, more particularly, those embodiments which are regarded at present as having preference, for example in connection with the preparation of powders of metal compounds such as uranium carbide (it being understood that the invention can be employed in obtaining other powders such as uranium nitride or uranium silicide), operation takes place as indicated below or in an analogous manner.

Such powders are desired at present particularly in the nuclear energy field, especially in the preparation of dispersions in inert matrixes, such as graphite or metal, or in the preparation of agglomerates by compaction. In these uses, it is desirable to employ substantially spherical particles and rigorously controlled dimensions, for example a diameter of 100 microns. Since at present it is hardly possible to obtain them otherwise than by grinding uranium carbide or the like, previously formed by sintering or fusion, the grinding step represents a considerably energy consumption and does not readily lead to the desired particle size range.

In carrying out the invention, use is made of the step of disintegration of a metal-ceramic composition, that is a metallic ceramic containing the refractory metal compound to be obtained, for instance uranium carbide, and a metal alone or alloyed, namely in this instance uranium, i.e. the base metal of the compound, the disintegration taking place by reason of the chemical or physical action of an external agent (such as a reactant or solvent) and

undergoing transformation into powders, from which the desired metal compound is extracted, e.g. by physical or chemical separation. In other words, the reaction in question gives a mixture in powder form containing both the residue from the reaction with or the dissolution of the metal and also the desired powder, which is separated out.

Such a process avoids any mechanical grinding treatment. It permits a good particle size to be obtained, particularly if the process includes, as is preferable, a heat treatment for modifying the grain structure prior to disintegration.

It is to be understood that the proportions of the metal compound and of the metal in the initial metal-ceramic composition are desirably chosen in the appropriate manner. Proportions in the range of 60 to 95% for the former and 40 to 5% for the latter are considered appropriate, though these amounts are not to be taken as limiting.

Assuming that it is desired to prepare uranium carbide powder, operation takes place as follows:

There is first prepared, by one of the known methods, for example sintering a "cermet" or metal-ceramic composition containing uranium monocarbide and uranium metal in the afore-mentioned proportions, which correspond to a carbon content of the composition in the range of 3 to 4.4% by weight. The carbon content of uranium monocarbide, UC, is 4.8%, from which it will be seen that a proportion of 3 to 4.4% of carbon in the composition gives a major amount of UC and a minor amount of uncombined uranium metal.

In preparing the metal-ceramic composition, pure uranium and pure carbon are used, for example, in the aforementioned proportions, and are subjected to sintering. By an alternative known method, uranium oxide is used and is reduced by carbon.

This composition is subjected to a heat treatment for grain-refining or enlarging the particles of uranium carbide, regularising their shape and making their size uniform.

The heat treatment is effected, for instance, under a vacuum of about 10^{-4} to 10^{-6} mm. of mercury or in an inert atmosphere for a period of 1 to 8 hours at a temperature in the range of 1200° to 2200° C.

After the heat treatment, the disintegration step is effected, for instance by heating the metal-ceramic composition in the presence by hydrogen, under conditions conducive to the formation of uranium hydride.

This treatment is carried out in a suitable furnace, for example at a temperature in the range of 200° to 300° C. and under a hydrogen pressure of about 1 atmosphere.

Depending upon the conditions of treatment, a mixture of uranium monocarbide and uranium hydride powders forms. The mixture then has to be separated into its constituents.

The separation is effected for example by decantation in a liquid of appropriate density and viscosity or in any other manner, e.g. by centrifuging.

It is to be understood that this separation may be effected directly on the uranium monocarbide and hydride derived from the afore-mentioned treatment or on a mixture of uranium carbide and uranium powder, after decomposition of the uranium hydride.

In the latter case, decomposition of the hydride takes place, for example, by heating under vacuum, at a temperature of about 400° C., the vacuum being obtained by a high-throughput pump.

The above-described or an analogous process can be used for the preparation of many kinds of powder, such as uranium nitride powders from a uranium nitride/uranium metal-ceramic composition, and uranium silicide (U_3Si_2) powders from a metal-ceramic composition composed

of the compound per se and the eutectic which it forms with uranium.

In the case of uranium nitride powders, disintegration occurs, for instance, by the action on the uranium nitride/uranium composition of normal hydrochloric acid, at a temperature of about 60° C., for example, thus producing uranium chloride which goes into solution. The uranium nitride powder forms in this solution. The reaction product is then subjected to filtration and the filtrate, namely the nitride powder, is washed with water, dried with alcohol or acetone and then dried under vacuum.

In the case of uranium silicide, action on the eutectic by one of the agents mentioned above for the carbide or the nitride or by some other agent causes decomposition into small particles, while the uranium silicide assumes the form of larger particles. It is thus appropriate to separate the two kinds of particles physically.

It may be mentioned that, in the case of the silicide, the grain-refining heat treatment should preferably be limited to temperatures in the range of 1000° to 1500° C.

Depending upon the particular procedure adopted, it is possible to prepare powders in a much more simple manner than at present, particularly because the grinding step, involving a high energy consumption, can be avoided.

As can be seen and as appears from the foregoing, the invention is not limited to the embodiments described nor to the ways of carrying out its various features especially envisaged above; it includes, on the contrary, all possible modifications.

What we claim is:

1. A process for the production of powders of refractory uranium compounds selected from the group consisting of uranium carbide, uranium nitride and uranium silicide, which comprises the steps of forming a mass of a metal-ceramic composition containing a major proportion of said compound and a minor proportion of uncombined uranium metal as the matrix metal of said mass, heating said composition at a temperature of from 1000° C. to 2200° C. for a time sufficient to regularize the shape and make uniform the size of the grains of said refractory uranium compound, forming particles of said compound by contacting said mass with at least one substance which chemically and selectively reacts with said uncombined matrix metal with disintegration of the mass into small particles of said refractory uranium compound and separating the particles of refractory uranium compound.

2. The process according to claim **1** in which said mass is disintegrated into substantially spherical particles of refractory uranium compound having a diameter of about 100 microns.

3. A process according to claim **1**, in which the metal-ceramic composition is formed by sintering the uranium compound and the uranium metal together.

4. A process according to claim **1**, in which the compound in powder form is separated mechanically from the other products produced by the action of the substance upon the metal.

5. A process according to claim **1**, in which the compound in powder form is separated physically from the other products produced by the action of the substance upon the metal.

6. A process according to claim **1** in which a metal-ceramic composition containing uranium nitride and uranium metal is contacted with hydrochloric acid which chemically reacts with the uranium metal to form uranium nitride powder in solution and the uranium nitride powder is separated by filtration.

7. A process according to claim **1** in which the substance which reacts chemically with the metal is selected from the group consisting of hydrogen and hydrochloric acid.

8. A process according to claim **1** in which the metal-ceramic composition initially contains 60 to 95% refractory uranium compound and 40 to 5% uranium metal.

9. A process according to claim **6** in which the treatment with hydrochloric acid is effected at about 60° C. with normal hydrochloric acid.

10. A process for the preparation of uranium carbide powder, which comprises forming a metal-ceramic composition containing uranium monocarbide and uranium metal and having a carbon content of 3 to 4.4% by weight, treating the composition with hydrogen to form uranium hydride by chemical reaction with the uranium metal and separating the desired uranium carbide powder from the other products of the reaction by decantation in a liquid.

11. A process according to claim **10**, in which the composition of uranium monocarbide and uranium is subjected to a grain-refining heat treatment at a temperature of 1200° to 2200° C. for a period of 1 to 8 hours.

12. A process according to claim **10** in which the treatment with hydrogen is carried out at 200° to 300° C. under a hydrogen pressure of about one atmosphere.

13. A process according to claim **11** in which the grain refining treatment is carried out under vacuum of about 10^{-4} to 10^{-6} mm. of mercury.

14. A process for the preparation of uranium silicide powder, which comprises forming a metal-ceramic composition containing uranium silicide and its eutectic with uranium metal, treating the composition with an agent selected from the group consisting of hydrogen and hydrochloric acid to effect chemical reaction with the uranium metal and cause the formation of particles of uranium silicide and physically separating the uranium silicide particles from the other reaction products.

15. A process according to claim **14**, in which the composition of uranium silicide and its eutectic with uranium metal is subjected to a grain-refining heat treatment at 1000° to 1500° C.

References Cited by the Examiner

UNITED STATES PATENTS

1,488,311	3/1924	Brockbank	23—208
2,158,520	5/1939	Meyer	23—208
2,485,175	10/1949	Trapp.	
2,534,676	12/1950	Newton et al.	23—14.5
2,784,054	3/1957	Carter et al.	23—14.5
2,913,314	11/1959	Osthoff	23—208
2,926,071	2/1960	Alexander	23—191
2,982,619	5/1961	Long	23—14.5
3,034,889	5/1962	Spedding et al.	23—14.5

OTHER REFERENCES

Dubuisson et al.: "2nd U.N International Conference on Peaceful Uses of Atomic Energy," vol. 6, pp. 551–560 (1958).

Katz et al.: "The Chemistry of Uranium," page 240 (1951), McGraw-Hill Book Co., N.Y.C.

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