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PROCESS OF FABRICATION OF SINTERED COMPOUNDS BASED ON URANIUM AND PLUTONIUM

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9 Claims. (Cl. 252—301.1)

Uranium and plutonium in the pulverulent state can be compressed and sintered either alone or in the presence of other metallic constituents and can form a sintered alloy which can be used as a fuel element in a nuclear reactor. They can also be combined with another element, for example carbon, nitrogen or silicon, in order to form after heating a novel compound also capable of providing a nuclear fuel by a powder metallurgy method. The production of such sintered products implies that the two base metals are available in the form of powders.

In order to obtain the base metal powders, either the metal oxides are reduced by means of calcium or the hydrides are decomposed.

These methods of preparation have serious disadvantages however.

Although the method of reduction of uranium oxide with calcium is used at present, it cannot be advantageously employed for reducing plutonium oxide, because washing of the production slags with dilute acids is incompatible with the very high oxidizability of plutonium.

Also, although direct action of hydrogen on massive uranium and plutonium provides pulverulent hydrides which can be substituted for the metals in all the applications which are initiated by starting with hydrogen at a slightly elevated temperature, the use of hydrogen in the presence of plutonium is not very desirable because of the danger of explosion which is connected with the use of that gas. Also subsequent fabrication steps necessitates a very slow dehydrogenation stage.

It is known that uranium-plutonium alloys having plutonium contents from 15 to 80% are very fragile, because of the presence of a zeta phase. They normally have fissures at ordinary temperature and are extremely friable. When stock-piled, they break down very rapidly.

The present invention consists in a process of fabrication of sintered compounds based on uranium and plutonium, the plutonium content of which is from 15 to 80%, in which, after provision of a uranium-plutonium alloy of the desired plutonium content by any known procedure, the alloy is ground in order to obtain a pulverulent product which is then compressed and sintered with the addition of another element or substance.

Metals which can be added to the pulverulent alloy include: molybdenum, niobium, tantalum and zirconium.

Nonmetallic elements and substances which can be combined with the alloy include carbon, silicon, nitrogen and hydrogen sulphide.

The process according to the invention can be put into effect very readily. It is sufficient in fact to grind the alloy directly, under an inert atmosphere, in a ball mill, for example. The powders obtained by pulverization of the alloy have an ability to become sintered which is much better than that of mixtures of uranium and plutonium powders obtained by standard processes, for example by calciothermy or by decomposition of the hydrides. They also have the advantage in the case where it is desired to make a mixed compound with a nonmetallic element of conducing directly to the formation of solid solutions of uranium and plutonium compounds. Production of the mixed compound can be effected either in a single stage, that is to say the grinding and reaction

of the alloy with the nonmetallic composition are simultaneous, or in two stages, in which preliminary grinding of the alloy in an inert atmosphere is then followed by reaction of the alloy with the nonmetallic compound. The preparation of other refractory fuel products from these powders, such as carbides for example, is more readily put into operation than by the reduction of oxides by carbon or carburization of hydrides.

Various examples are given below, by way of illustration only, of the process of manufacture of sintered compounds based on uranium and plutonium in accordance with the invention.

Example 1

This example relates to the preparation of a nuclear fuel based upon uranium and plutonium carbides.

(a) A uranium-plutonium alloy, the plutonium content of which is 30%, is prepared in an arc furnace;

(b) The alloy obtained is ground in a ball mill under an atmosphere of argon for 10 minutes in the presence of the theoretical quantity of carbon for the production of the carbides; the average grain diameter is then less than 40μ;

(c) The powder obtained previously tabletted under a pressure of 7.25×10^{-2} p.s.i., in order to effect the carburization reaction, is then heated under vacuum at 1200° C. for 10 hours;

(d) the carbide obtained is ground in a ball mill for 20 minutes in the presence of 1% of naphthalene and 0.5% of nickel by weight; the average grain diameter is then less than 40μ;

(e) The carbide powder and additive are compressed under a pressure of 7.25×10^{-2} p.s.i.; the apparent density obtained is 9.2 gm./cc.;

(f) The tablets obtained are sintered under vacuum at 1450° C. for 4 hours; the refractory fuel material so obtained has a density of 13.45 gm./cc.

Example 2

This example deals with the preparation of plutonium and uranium mononitride.

(a) A uranium-plutonium alloy having a plutonium content of 30% is prepared in an arc furnace;

(b) The alloy obtained is ground in a ball mill for 5 minutes under an atmosphere of argon;

(c) After introducing the stoichiometric quantity of nitrogen to form the mononitrides, the mixture is heated for 1 hour at 350° C.;

(d) The powder is then sintered by any known means.

I claim:

1. A process for the production of a sintered uranium-plutonium product comprising the steps of grinding to a powder, fragile particles of a uranium-plutonium alloy, the plutonium content of which is from 15 to 80%, and compounding said uranium-plutonium powder by compressing and sintering the ground powder in the presence of compounding metallic and nonmetallic materials selected from the group consisting of molybdenum, niobium, tantalum, zirconium, carbon, silicon, nitrogen and hydrogen sulphide.

2. A process according to claim 1 in which the compounding material is carbon and a carbide of said uranium-plutonium alloy is formed.

3. A process according to claim 1 in which the compounding material is nitrogen and a nitride of said uranium-plutonium alloy is formed.

4. A process according to claim 1 in which the grinding is conducted in an inert atmosphere.

5. A process according to claim 1 in which the compounding material is added during the grinding.

6. A process according to claim 1 in which the compounding material is added to the ground powder.

7. A sintered uranium and plutonium product prepared according to the process of claim 1.

8. A process for the production of an uranium-plutonium carbide nuclear fuel comprising the steps of grinding under an inert atmosphere fragile particles of a zeta phase containing uranium-plutonium alloy, the plutonium content of which is 30%, in the presence of the stoichiometric quantity of carbon to provide a 40μ grained powder, compressing this powder at a pressure of about 7.25×10^{-2} p.s.i. into a compact, heating said compact under vacuum at 1200° C. to effect carburization, grinding the carburized product, compressing the carburized powder and sintering at 1450° C.

9. A process for the production of an uranium-plutonium nitride comprising the steps of grinding a fragile zeta phase containing alloy of uranium and 30% plutonium in an inert atmosphere, introducing a stoichiometric quantity of nitrogen and heating to 350° C. to form nitrides and then sintering the nitrided powder.

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