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2,946,699

PROCESS OF IMPREGNATING GRAPHITE WITH A URANIUM COMPOUND

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No Drawing. Filed Mar. 24, 1947, Ser. No. 736,626 4 Claims. (Cl. 117-65)

This invention relates to atomic energy adapted for 15 use in rocket propulsion, as well as nuclear chain reaction power sources generally.

It is an object of this invention to provide a new and novel arrangement of a body material embodying an active substance. (Active substance is defined as material capable of supporting nuclear chain reactions, including such materials as uranium, plutonium, and the like, as well as compounds of any of these materials. The active substance is carried by a body material which may serve only as a support for the active substance or 25 as a moderator and support. In the case where the material serves only to support the active substance in such a way that the heat transfer can be effective, the reactor is known as a fast neutron reactor. Where the material resonance neutron reactor, or a slow neutron reactor.)

It is another object of this invention to provide a moderator which effectively decreases the velocity of the neutrons involved in the nuclear chain reaction without appreciably absorbing them.

It is a further object of this invention to provide a new and novel process for impregnating a porous material with an active substance.

It is yet another object of this invention to provide a process which will result in a high ratio of impreg- 40 nation of porous material with active substance.

It is yet another object of this invention to provide a process which will result in thorough and uniform impregnation of the porous material by the active substance.

It is still another object of this invention to impregnate the porous material with a salt of the active substance in such a manner that the salt provides the oxygen for obtaining the final form of the relatively stable active substance.

It is yet another object of this invention to impregnate a porous substance with a solution of a salt of an active substance in such a manner that a solution of high concentration is obtained by dissolving or melting it in its own water of crystallization.

Further objects of the invention will become apparent from the disclosure herein contained.

In carrying out the present invention the porous substance is impregnated by a vacuum-pressure process utilizing a compound or salt of a fissionable material, such as uranyl nitrate hexahydrate, UO2(NO3)2.6H2O. The salt may obviously be composed of fissionable and non-fissionable material in desired proportions depending upon results desired. This salt is dissolved or melted in its own water of crystallization by heating at a temperature above 60° C. and then, after the impregnation, it is oxidized by heating to a temperature that will result in the formation of uranium trioxide (UO3) or uranium oxide (UO2). This is accomplished in the following manner:

Graphite is placed in a container containing uranyl nitrate hexahydrate. The container may then be evacu-

ated to exhaust the air therefrom as well as from the porous graphite and thus facilitate impregnation of the graphite with the melted salt, although satisfactory results may be obtained by pressurization only. In any event, the atmosphere within the container is controlled according to results desired. The container is then sealed and heated to approximately 130° C. for a sufficient period of time to cause the salt to melt and be received in the graphite. (The salt melts at approximately 60° C. under normal conditions and at higher temperatures under evacuated conditions.) Continued heating results in the establishing of a pressure condition within the container. When the pressure condition has been continued a sufficient length of time to thoroughly impregnate the graphite with the salt, the impregnated graphite is removed from the container and first heated to drive off the water from the hydrous impregnated salt and then further heated to drive off NO2 gas to convert the impregnated material to UO3 or UO2. It has been found that heating slowly to approximately 275° C. will result in conversion to UO3 while heating to higher temperatures will result in other forms of uranium oxide. For instance, UO2 will form at approximately 1400° C. In this embodiment of the invention, .2 to :25 gram of uranium metal has been included in each cc. of material of 25% porosity while approximately .4 to .5 gram of uranium has been included in material of approximately 40% porosity.

In another embodiment of the invention, the porous serves also as a moderator, the reactor is known as a 30 material may be impregnated in a vacuum-pressure process using the anhydrous uranyl nitrate, UO2(NO3)2, and oxidizing the impregnated salt in one continuous process. As in the case of the uranyl nitrate hexahydrate, the process is finally carried on at a temperature of approxi-35 mately 275° C. to form UO3, although decomposition to the UO3 state may take place at some lower temperature. The process may be carried on at higher temperatures to obtain uranium oxide in different forms, UO₂ being formed at approximately 1400° C.

In carrying out the process continuously, the graphite is placed in a container adapted to be evacuated and/or pressurized. UO₂(NO₃)₂6H₂O is placed in the container which may then be evacuated or pressurized, as desired, and heated until substantially all the water is driven off from the hydrous salt. A certain percentage of water is not objectionable and may serve to assist in the melting of the salt, since it will be driven off in any event at a later stage of the process. The container is then suitably pressurized, according to the result desired, to cause the resulting melted salt to be received within the graphite. Heating is then continued slowly up to 275° C. to break down the anhydrous salt and form UO3. (The minimum temperature for the oxide formation is approximately 235° C.) NO₂ is driven off in the process, the container being suitably vented to the atmosphere to maintain the desired pressure within the system. The heating is continued until NO2 gas ceases to be given off, this being an indication that the process is complete. At this time the pressure will drop within the container and the reaction substantially cease.

In this embodiment of the invention, approximately .45 gram of uranium has been embodied in each cc. of material of 35% porosity larger amounts being received in material of greater porosity. This form of the invention has certain advantages over the first described form in that by oxidizing the salt in one continuous process the salt is prevented from being extruded from the pores of the material upon cooling and subsequent reheating of the moderator or nuclear reactor.

The nuclear reactor may be formed by compounding active substance with the body material which carries the active substance or for use as a moderator in connection therewith. For instance, powdered graphite may be mixed with an active substance, such as a metal, metallic oxide, or metallic carbide, and then subjected to temperature and pressure conditions to form a nuclear reactor. The materials are combined, of course, in proportions 5 depending upon results desired. The resulting compound may be obtained by sintering the body material and the active substance, or combining the porous substance with the molten metal or molten carbide of the metal. This procedure facilitates the impregnation of 10 is of approximately 25% porosity, and in which the the body material in varying amounts so that reactor material of different proportions of active substance and moderator or body material may be obtained for use in different parts of the pile. In order to obtain a highly condensed reactor, the same may be formed by com- 15 bining the active substance and body material under high temperature conditions and pressure conditions such as may be obtained by a ram. A reactor so formed is particularly adapted for use at high temperatures and locations where very high concentrations of fissionable 20

material is desired. Although the invention has been described in detail, it is to be clearly understood that the same is by way of example only, and is not to be taken by way of limitation, the spirit and scope of this invention being 25 limited only by the terms of the appended claims.

We claim:

1. A process of forming reactor material comprising the steps of placing in a container uranyl nitrate hexahydrate, and graphite; heating the uranyl nitrate hexahydrate and graphite to a temperature greater than about 60° C. to melt the uranyl nitrate hexahydrate in its own water of crystallization so that it will then be absorbed by the graphite; and thereupon heating the graphite with the salt absorbed therein to a temperature greater 35 than about 235° C. until the salt is converted into an

2. A process as recited in claim 1 in which the uranyl nitrate hexahydrate and graphite are heated to a temperature between 60° C. and 130° C. and the graphite, 40 with the uranyl nitrate hexahydrate absorbed therein, is heated to a temperature above 235° C. and in the neigh-

4 borhood of 270° C. to convert the salt to uranium tri-

3. A process as recited in claim 1 in which the graphite is of approximately 40% porosity and in which the graphite, with the uranyl nitrate hexahydrate salt therein, is heated to approximately 275° C. to embody in the graphite approximately 0.4 gram of uranium per cc. of

4. A process as recited in claim 1 in which the graphite graphite, with the uranyl nitrate hexahydrate salt therein, is heated to approximately 275° C. to embody in the graphite approximately 0.2 gram of uranium per cc. of graphite.

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