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[54] METHOD OF MAKING NUCLEAR FUEL-CONTAINING BODIES

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[57] ABSTRACT

In the forming of bodies, containing fission product retaining nuclear fuel particles, dispersed in a filler material, it is desirable to produce a compact and coherent body without any fractured fuel particles. Fractured particles occur during pressing and the risk of fracture may be reduced by overcoating the fuel particles with two layers of material which exhibit during pressing different rheological characteristics. The characteristics differ in that the inner layer is less readily deformable under the pressure than the outer layer which tends to flow into the spaces between the overcoated particles. On such deformation of the outer overcoating, the inner overcoating layer acts as a buffer between adjacent particles.

14 Claims, No Drawings

METHOD OF MAKING NUCLEAR FUEL-CONTAINING BODIES

BACKGROUND OF THE INVENTION

This invention relates to the manufacture of nuclear fuel containing bodies.

One method of employing fission product retaining fuel in nuclear reactors which has been proposed hitherto is to take a batch of fission product retaining nuclear fuel particles which are fuel kernels already coated with their fission product retaining coating and to apply to each of these an overcoating of an adherent but deformable character, for example, a mixture of synthetic polymerizable resin and graphite powder. A mass of coated particles, so overcoated, is then placed in die and pressed into a coherent fuel containing body which may be readily handled as a fuel component.

Although it might appear that equally sized particles uniformly overcoated, may, on pressing, result in a uniform distribution of particles within the body, in practice the pressing operation generates non-uniform pressures and asymetric flow of the overcoating material (which forms ultimately the matrix), generating a risk of inter-particle contact which could lead to coating fracture. Even if inter-particle contact does not occur, an irregular distribution of fuel particles within the compact may result.

It will be appreciated by those skilled in the art that an irregular distribution of fuel within the artefact is undesirable not only by the risk of particle damage but also from heat transfer and reactor physics points of view.

SUMMARY OF THE INVENTION

According to the invention a pressed nuclear fuel containing body comprises a coherent mass of fission product retaining fuel particles overcoated with at least two overcoating layers of which the outermost layer, having been formed of more deformable filler than the layer nearer to the fuel particle, has substantially filled, on pressing, the interstices between the overcoated particles, the coated particles being separated by the inner layer of overcoating.

The overcoating layers are of such a nature that they exhibit different characteristics during the pressing operation to enable them to serve the following functions. The inner overcoating layer is a substantially firm layer and prevents the underlying fission product retaining particles from coming into contact. The outermost overcoating layer is preferably of a thickness sufficient to fill voids between a mass of the particles i.e. it represents approximately 35 percent or more of the intended final volume of the body, and its viscosity is such that, on the application of pressures used in these processes, it will flow into these voids or interstices.

The invention also resides in a method of making a nuclear fuel containing body as aforesaid which resides in the steps of applying at least two layers of overcoating material on to fission product retaining nuclear fuel particles, the rheological characteristics of the layer nearer to the fuel particle being different from those of the outer layer such that the outer layer deforms to a greater degree than the inner layer, placing a batch of such particles in a cavity and subjecting them to a pressing operation which causes the material of the outer layer substantial to fill the interstices between the particles whilst maintaining the particles separated by the less deformable inner layer of overcoating material.

A suitable difference in rheological characteristics of the layers of overcoating material may be brought about in a number of ways. For example, the composition of the material used for each layer may be different (EXAMPLE 2), the ingredients of each layer may be the same but their proportions may vary (EXAMPLE 1), the layers once formed may be subjected to different physical treatment at an intermediate stage between the two coating stages (EXAMPLE 3), or a change may be made in the sources of the material included in the layers or by using different grain size of this material such that its rheological characteristics differ(EXAMPLE 4).

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The following examples are demonstrative of the invention.

EXAMPLE 1

A quantity of substantially spherical kernels made from UO_2 grains and an aluminum stearate binder were coated with three layers of pyrolytic carbon silicon carbide and pyrolytic carbon in that order to produce a fission product retaining fuel particle. The particles were graded to produce batches of coated particles substantially uniform overall diameter. The overcoating process which consisted in the application of two layers of overcoating material was carried out as follows.

1. Preparation of the Overcoating Material

A quantity of calcined petroleum coke was crushed and sieved to provide a fraction below 50 microns and this fraction was graphitized by heating to 2,600°C. The graphitized powder when cold was mixed with a solution containing a predetermined quantity of phenol formaldehyde resin. The solvent was evaporated and the coke crushed to provide a fraction below 80 microns of graphite grains incorporated with resin. The quantity of resin was about 12 percent (by wt) of the grains. This substance is referred to herein as overcoating material 12 percent polymerizable resin.

A further quantity of crushed calcined petroleum coke was similarly treated but with the difference that the final resin content of the coated grains was 25 percent. This substance is referred to herein as overcoating material 25 percent polymerizable resin.

2. The Application of the Overcoating Material

A batch of fission product retaining particles was placed in a drum which had one open end and was 2 feet diameter. The drum was mounted with its axis inclined at 15° to the horizontal and was rotated at about 30 revolutions per minute. A supply of overcoating material 12 percent resin was allowed to trickle under gravity into the drum and the grains were sprayed intermittently with methylated spirit, a solvent for the resin, as the grains entered the drum. The resin binder content of the grains was thus softened and these grains then adhered to the tumbling particles in the drum. The process was continued until an overcoating layer of 50 μ m had been built up. The rotation of the drum was then stopped and also the supply of overcoating material 12 percent resin.

If necessary the partially overcoated particles may be sieved at this point to ensure a uniform diameter and returned to the drum.

The overcoating process was resumed with the supply of overcoating material previously used replaced by a supply of overcoating material 25 percent resin. The second overcoating layer was applied in a manner similar to the first. Again, when a layer about 50 m thick had been applied the process was stopped and the particles removed from the drum.

The batch of particles bearing their double layers of overcoating was then placed in a cylindrical die cavity and brought to a temperature at which the resin content of the overcoating softened and yet did not undergo unduly rapid polymerization, in this case about 80°–90C. A plunger was used to apply a pressure of about 1,000 psi to the particles so that the mass assumed a shape conforming to the die cavity. The outer layer of overcoating material flowed to substantially fill the interstices between the particles while the inner layer(s) remained substantially undeformed ensuring that the surfaces of the fission product retaining coatings on adjacent particles were maintained separated by a thickness of matrix equal to about twice the thickness of the inner less deformable overcoating layer. When the full pressure had been applied and movement of the plunger and die contents had ceased, the temperature of the die was raised to 180°C so that the resin polymerized throughout the body. The body was then ejected and heated to 800°C over several hours in an inert atmosphere to carbonize the resin and on examination the body was found to be a coherent mass of fuel particles dispersed, quite uniformly, in a matrix of graphite.

EXAMPLE 2

This example followed example 1 with the exception that material employed for both overcoating layers had the same resin content but a quantity of lubricant/plasticizer e.g. stearic acid was incorporated in the outer layer material (note stearic acid melts at 69°C and therefore liquifies and lubricates at the pressing temperature) thus promoting preferential deformability in the outer layer during the pressing operation. Otherwise the process parameters were identical with those of example 1.

EXAMPLE 3

In this example the material used for both layers of the overcoating is of identical composition. However, on completing the first layer the temperature of the batch is raised to partly, or completely, polymerize the resin of the first layer after which the second layer is applied. The polymerizing process may be carried out in the overcoating drum or in a separate vessel. The pressing operation takes place as before; only the unpolymerized outer layer being capable of ready deformation. Complete polymerization of both layers is then carried out under pressure as before.

As a modification in example 3 the partial, or full, polymerization of the inner layer may be achieved by contacting the particles between coating stages with a polymerizing agent.

EXAMPLE 4

It has been found that graphite powders of different size distribution, when unimpregnated with identical amounts of resin, have very different rheological characteristics when employed pressing operations such as previously described.

In this example, the differing deformabilities of the component layers of the overcoating is achieved by the use of two graphite powders of different origin, or of different particle size distribution, both having been impregnated with the same amounts of resin and with similar amounts of plasticizer and/or lubricants.

For example, the use of powder produced by grinding a Gilsonite based graphite stock and impregnated with 12 percent resin for the inner layer forms an overcoating layer which is less deformable in the pressing process than an outer layer using a graphitized needle coke powder containing the same percentage of resin.

As a modification the graphite of similar type and origin can be used in both layers but the respective powders varying only in particle size and particle size distribution, again having the effect of promoting more ready deformability in the outer layer.

We claim:

1. In a method of making a solid nuclear fuel-containing body composed of fission product-retaining nuclear fuel particles wherein fuel kernels having a fission product-retaining coating are provided with an overcoating of an adherent but deformable character and a batch of the overcoated particles are placed in a cavity with the overcoatings of adjacent particles in contact and compacted under pressure into a coherent fuel-containing body which may be readily handled as a fuel component, the improvement wherein said overcoating is provided in distinct inner and outer layers, both of said layers being provided before subjecting the particles to pressure, the rheological characteristics of the overcoating layer nearer to the fuel particle being different from those of the outer overcoating layer such that the outer overcoating layer deforms to a greater degree than the inner overcoating layer whereby, on compacting the particles under pressure, the material of the outer overcoating layer fills the interstices between particles while maintaining the particles separated by the less deformable inner overcoating layer.

2. A method according to claim 1 wherein said inner and outer overcoating layers each comprise carbon particles and resin.

3. A method of making a body as claimed in claim 2 in which the differences in rheological characteristics are brought about by providing different proportions of the ingredients in the two overcoating layers.

4. A method as claimed in claim 2 in which the material of the overcoating layers comprises graphite powder impregnated with polymerizable resin, the material for the outer layer containing a higher proportion of resin than that for the inner layer.

5. A method as claimed in claim 2 in which the difference in rheological characteristics are brought about by providing different compositions of materials of the two overcoating layers.

6. A method as claimed in claim 5 in which the composition of the material for the outer layer includes a pressing lubricant or plasticizer.

7. A method as claimed in claim 2 in which the differences in rheological characteristics are brought about by providing both different ingredients and different proportions of ingredients in the inner and outer layers.

8. A method as claimed in claim 2 in which material for both inner and outer layers include a polymerizable resin, the inner layer being at least partially polymerized before the application of the outer layer.

9. A method as claimed in claim 8 in which the fission product retaining particles are coated with an inner overcoating layer of graphite powder and polymerizable resin, the inner layer being at least partially polymerized prior to the application of the second layer of similar overcoating material, the particles being pressed with the outer layer in the unpolymerized condition and then heated while the pressure is maintained to complete polymerization of the whole.

10. A method of making a body as claimed in claim 2 in which the overcoating materials comprises powdered graphite and polymerizable resin and the different rheological characteristics of the layers is brought about by the selection of the graphite.

11. A method of making a body as claimed in claim 10 in which the material for the inner layer of overcoating includes Gilsonite graphite powder and polymerizable resin and the material for the outer layer of overcoating includes graphite powder, derived from needle coke, and polymerizable resin.

12. A method of making a body as claimed in claim 2 in which the overcoating materials for the inner and outer layers comprise graphite powders of similar origin but of different grain size.

13. A method of making a body as claimed in claim 2 in which the material for the outer layer contains a higher proportion of pressing lubricant than the material for the outer overcoating.

14. A method of making a pressed body comprising a mass of fission product retaining nuclear fuel particles dispersed in a matrix of graphitic material which process comprises applying to each particle an inner overcoating comprising powdered graphite and polymerizable resin, applying a second similar coating but having a higher resin content than the inner layer of a character which is more easily deformable under pressure than the inner layer, placing the overcoated particles in a die cavity, applying pressure to the particles while the die content is heated to soften the resin and than after pressing has deformed the outer layer of overcoating, polymerizing the whole of the unpolymerized resin while the pressure is maintained.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,708,559 Dated January 2, 1973

Inventor(s) Eric H. Voice; Leslie W. Graham; Derek W.J. Sturge

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Claims priority of British Application Serial No. 14023 filed March 23, 1970.

Signed and sealed this 29th day of May 1973.

(SEAL)
Attest:

EDWARD M.FLETCHER,JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents

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