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MULTILAYERED NUCLEAR FUEL ELEMENT

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BACKGROUND OF THE INVENTION

The present invention relates to nuclear fuels and methods of their manufacture, and, more particularly, to nuclear fuel elements that retain their physical integrity in high temperature gas reactors (HTGR) where normal operating temperatures may exceed the melting temperatures of the fissionable nuclear material contained within the fuel elements.
To realize an efficient nuclear propulsion system, the nuclear reactor that powers the system must be small and light-weight and yet be able to provide power at a substantially high output rate. HTGR's have been seen as an effective means of attaining the requisite power densities needed to power such propulsion systems. To obtain such high power outputs, however, HTGR's must operate at very high temperatures. Yet, due to structural constraints of the fuel elements, most prior art HTGR's have been designed to operate at temperatures that normally do not exceed about 1,500° C and not in excess of the melting point of the fissile fuel material contained in the nuclear fuel elements.

As noted above, the primary limitation to high temperature operation of the HTGR's have been the nuclear fuel elements which fail at temperatures above the melting point of the fissile material that is contained within the fuel elements. Failure of nuclear fuel elements at high temperatures results from outward migration of the molten fissile material and decomposition and/or reaction of the carbide coating surrounding the fuel. At temperatures above the melting point of the fissile material, the molten fuel migrates outwardly and attacks the coating of the fuel element and causes release of fuel and fission products into the surrounding reactor environment. At temperatures significantly above 2000° C, the carbide materials which typically make up the outer coating decompose and fail or become nonstoichiometric.

What is needed is a fuel element that can withstand the pressures and temperatures that may be generated in a HTGR operating at a temperature
substantially beyond 2000° C. To prevent undesired release of fissionable material into the reactor environment, the fuel element must be able to retain its physical integrity and the fissile material at temperatures substantially beyond the melting point of the fissile material.

In view of the foregoing, the general object of this invention is to provide a nuclear fuel element that can operate at temperatures substantially beyond the melting point of the fissile material contained within the fuel element.

Another object of this invention is to provide a nuclear fuel element having a multilayered structure which effectively retains the fissile material within the layered structure while protecting the fuel element from the moderating gases found in a HTGR which is operating at high temperatures.

Yet another object of this invention is to provide a method of constructing a multilayered nuclear fuel element that can operate at temperatures substantially beyond the melting point of the fissile material contained within the fuel element without failing.

Additional objects, advantages and novel features of the invention will become apparent to those skilled in the art upon examination of the following and by practice of the invention.

**SUMMARY OF THE INVENTION**

To achieve the foregoing and other objects, this invention provides a nuclear fuel element suitable for high temperature applications such as in high temperature gas reactors. The fuel element includes a kernel of fissile material which is overlaid
with concentric layers of impervious graphite, vitreous carbon, pyrolytic carbon and metal carbide. The kernel of fissile material is surrounded by a layer of impervious graphite. A layer of vitreous carbon then surrounds the layer of impervious graphite. Finally, an outer shell which includes alternating layers of pyrolytic carbon and metal carbide surrounds the layer of vitreous carbon.

**BRIEF DESCRIPTION OF THE DRAWINGS**

The present invention is illustrated in the accompanying drawings where:

FIG. 1 is a cross-sectional view of a nuclear fuel element showing a kernel of fissile material surrounded by multiple concentric layers of coatings.

**DESCRIPTION OF THE PREFERRED EMBODIMENT**

The invention as described herein provides for a nuclear fuel element 10 suitable for high temperature applications such as in high temperature gas reactors (HTGR). The fuel element 10 includes a kernel of fissile material 11 which is overlaid with concentric layers of impervious graphite 12, vitreous carbon 13, pyrolytic carbon 14 and metal carbide 15. The multilayered structure of the fuel element 10 serves to maintain the physical integrity of the fuel element 10 at high temperatures by preventing the outward migration of molten fissile material 11, while protecting the fuel element 10 from reaction and decomposition from contact with moderating gases that are within a nuclear reactor. As shown in FIG. 1, the fuel element 10 is constructed of an inner kernel of fissile material 11 which is suitable for sustained nuclear fission reactions. In the preferred embodiment, the kernel of fissile material 11 is composed of uranium carbide, but the fissile material 11 need
not be limited to such and may comprise a number of various uranium compounds which are advantageous for sustained fission reactions. A preferred method of forming the uranium carbide fissile material 11 is described in U.S. Patent No. 5,094,804 (Schweitzer), entitled "Nuclear Fuel Elements and Method of Making Same." The cited patent discloses a method of forming a stable nuclear fuel kernel by impregnating a porous graphite substance with a conventional solution of fissionable fuel and heating the graphite substance sufficiently to form uranium carbide.

Referring again to FIG. 1, the kernel of fissile material 11 is first coated with a layer of impervious graphite 12. The layer of impervious graphite 12 encapsulates the fissile material 11 and serves to impede the outer migration of the fissile material 11, even when the fissile material 11 is in a molten state. The impervious graphite 12, being of the lowest free energy allotropic form of carbon, is highly resistant to chemical attack from the fissile material 11, even at the typically high operating temperatures of the nuclear fuel element 10.

Next, after the kernel of fissile material 11 is coated with the layer of impervious graphite 12, a layer of vitreous, or glassy, carbon 13 is applied to the impervious graphite 12. The layer of vitreous carbon 13, which exhibits excellent compatibility with the impervious graphite 12, is coated so as to completely surround the impervious graphite 12. The vitreous carbon 13 is stable at high temperatures and possesses sufficiently low permeability such that the layer of vitreous carbon 13 forms a gas tight container which is capable of confining both the fissile material 11
and other highly volatile fission products that are produced in nuclear fission. Suitably, the layer of vitreous carbon 13, in conjunction with the outer layers of pyrolytic carbon 14 and metal carbide 15, forms a small but sturdy pressure vessel capable of withstanding, not only the large internal gas pressures that are developed by the formation of volatile fission products, but also the external forces that are generated between fuel elements 10 that are operating in a nuclear reactor such as the HTGR.

The vitreous carbon 13 considered herein refers to a class of carbons that are hard and brittle like glass with an amorphous glassy structure. The vitreous carbon 13 is impervious to gases, has extensive cross-linking, resists graphitization, and maintains its amorphous structure and imperviousness at high temperatures. The vitreous carbon 13 is formed by pyrolysis of infusible plastics in the solid phase from starting materials that have three dimensional cross-linked structures and produce high carbon residues. Suitable materials for producing vitreous carbon from pyrolysis in the solid phase are phenolic and furan resins, polyphenylenes, polymides, and aromatic epoxy materials.

In the preferred embodiment, the layer of vitreous carbon 13 is applied by coating the impervious graphite layer 12, by means of dipping, spraying, or immersion, with a suitable material as described above and processing the coated fuel element 10 into small spheres by methods commonly used in the plastics industry. Pyrolysis can be carried out at varied temperatures, pressures, and times to obtain the vitreous carbon 13 with the desired physical characteristics. Vitreous
carbons 13 with excellent properties have been produced by pyrolysis temperatures between 1000° C and 3000° C.

The vitreous carbon 13 of the preferred embodiment has significantly different properties from pyrolytic carbons, including isotropic pyrolytic carbons. Unlike the vitreous carbons, pyrolytic carbons are normally formed from the cracking of organic gases and produced in fluidized and spouted beds. Further, pyrolytic carbons are much more permeable to gases than vitreous carbons and exhibit a wider range of structural characteristics as well as porosities. In general, the physical properties of vitreous carbons are more readily reproduced than pyrolytic carbons.

Lastly, the layer of vitreous carbon 13 is coated with alternating layers of pyrolytic carbon 14 and metal carbide 15, to form a rigid outer shell 16 which serves to protect the fuel element 10 from external forces such as mechanical and chemical effects of the moderating gases within a HTGR. In addition, the outer shell 16, in conjunction with the layer of impervious graphite 12 and vitreous carbon 13, as described above, forms a pressure vessel which is effective for retaining the fissile material 11 and fission products within the fuel element 10. In the preferred embodiment, as shown in FIG. 1, the invention provides for two alternating layers of pyrolytic carbon 14 and metal carbide 15 which are applied so as to completely surround the layer of vitreous carbon 13, as well as each subsequent layers. This layered structure also allows the inner layer of the metal carbide 15 to be maintained in a carbon rich environment and thus insure stoichiometric composition and low
evaporation rate of the metal from the metal carbide 15. The metal carbide 15 may include a variety of carbon compounds such as zirconium, niobium and tantalum.

In an alternate embodiment, one or both of the layer of pyrolytic carbon 14 may be substituted with vitreous carbon. This alternate structure using vitreous carbon may be appropriate when additional air-tightness is desired and/or when the chemical reactivity of the layer of metal carbide 15 is low.

The multilayered structure of the present invention is effective in significantly increasing the life of the fuel element 10. The multiple layers, not only reduce the possibility that a defect in just one of its multilayers will cause a failure of the entire fuel element 10, but also, significantly slows the rate of diffusion of the fissile material 11 and fission products, since their diffusion becomes inhibited at each of the layer interfaces. In addition, the multilayered structure allows the individual layers of the metal carbide 15 to be made much thinner than in a single layer structure. This advantage is important, since the number of defects which can cause cracks in the layer of metal carbide 10 becomes increasingly greater as the layer of metal carbide 15 is made thicker. It has been demonstrated that laminated layers, in contrast to single layers of equal thickness, are more effective in stopping crack propagation and in handling mechanical stresses.

Although the physical configuration of the nuclear fuel element 10 may vary in shape and size in accordance with the application desired, in the preferred embodiment, the fuel element 10 is generally spherical in configuration with a diameter of about 500-1000 microns. This particular configuration has been shown
to be advantageous for applying the multilayered coatings of the present invention using a number of conventional techniques such as fluidized or static beds or by reaction with solutions. Additionally, this particular size and shape has been demonstrated to exhibit good heat transfer properties and to minimize the effects of both internal and external gas pressures. The preferred thickness of the individual coatings or layers as described herein is about 10-50 microns. At thicknesses greater than 50 microns, defects, which can cause cracking, become increasingly more significant.

The multilayered structure of the present invention may be constructed using a number of conventional techniques. These techniques include using conventional fluidized or static beds to form layers by chemical vapor deposition (CVD), chemical vapor infiltration (CVI) and chemical vapor reaction (CVR). Using these techniques, the desired coating type or thickness can be achieved by varying the factors such as deposition temperature and time, gas composition and flow rate, and start-up/shut-down attributes. An alternate method of forming the multilayered coating is by dipping or reacting the kernel of fissile material 11 with various solutions. In this method, the desired structural characteristics of the multilayered coating may be achieved by varying the factors such as temperature, solution concentration, and removal and reinsertion of the fuel element 10 in and from the solution.

The foregoing description of a preferred embodiment of the invention has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the invention to the precise form disclosed. For example, the
alternating layers of pyrolytic carbon 14 and metal carbide 15 need not be limited to the two coating as described herein. The numbers of these layers may be increased or decreased as necessary to adapt the fuel element 10 to its operating environment.

The embodiment described herein explains the principles of the invention so that others skilled in the art may practice the invention in various embodiments and with various modifications as suited to the particular use contemplated. It is intended that the scope of the invention be defined by the claims appended hereto.
ABSTRACT OF THE INVENTION

A nuclear fuel element suitable for high temperature applications comprised of a kernel of fissile material overlaid with concentric layers of impervious graphite, vitreous carbon, pyrolytic carbon and metal carbide. The kernel of fissile material is surrounded by a layer of impervious graphite. The layer of impervious graphite is then surrounded by a layer of vitreous carbon. Finally, an outer shell which includes alternating layers of pyrolytic carbon and metal carbide surrounds the layer of vitreous carbon.