Changes in Material Properties of PyC Coating Layers by Neutron Irradiation in HTR Coated Particle Fuel: I. Dimensional Change

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1. Introduction

It is well known that the TRISO(tri-isotropic)-coated fuel particle for a HTR(High Temperature gas-cooled Reactor) has a diameter of about 1 mm, composed of a nuclear fuel kernel and four different outer coating layers, consisting of a buffer PyC (pyrolytic carbon) layer, inner PyC layer, SiC layer, and outer PyC layer with different coating thicknesses following a specific fuel design.

The fuel kernel is a source for a heat generation by a nuclear fission of fissile uranium. The role of each of the four coating layers is different in view of retaining the generated fission products and other interactions during in-reactor service. [1]

Among these coating layers, PyC properties are scarcely in agreement among various investigators and the dependency of their changes upon the deposition condition is comparatively large due to their additional anisotropic properties.

While a recent review work [2] has contributed to a clear relationship between the material properties and QC measurements, an integration of the data and modeling on the irradiation behavior of PyC coating layers remains at the preliminary stage since the middle of 1970's for the early BISO design. In this work, an attempted was made to analyze the existing data of the experimental results of the material property change upon neutron irradiations of PyC coating layer obtained from various experiments performed in early periods of the HTR coated particle development by collecting and comparing the different data, mainly in the dimensional aspect.

2. Dependence of material properties of PyC coating on deposition conditions

The material properties of a PyC coating layer for coated particle fuels have been periodically reviewed [3-5] and their relevant models have been suggested during the 1990s for their use in the analyses of an in-reactor performance of materials. Recent review work [2] summarized the material properties depending on the deposition conditions. Fig. 1 shows, as an example, the deposition temperature-density relationship of PyC layer when deposited using the methane and propane as hydrocarbon source gases with various contents. [6-9]

Generally, for LTI-PyC using propane as a source gas, PyC coating density decreases with increasing deposition temperature, meanwhile for HTI-PyC using methane the density increases with the deposition temperature. On the other hand, Young's modulus decreases with increasing deposition temperature for both LTI- and HTI-PyC. These material properties depend basically on the structure to be obtained during the coating process.



Fig. 1. Variation of PyC coating density as a function of deposition temperature.

3. Density change of PyC coating layer by neutron irradiation

Density of PyC deposited by a fluidized bed CVD coating is an important parameter that must be controlled in order that particles behave satisfactorily during irradiation, as it affects and represents a number of mechanical and physical properties such as elastic constants and the thermal conductivity. Its current specifications used among several organizations range between 1.85 and 1.95 g/cm³ for dense PyC layers and 0.85 and 1.20 g/cm³ for buffer layer [10]. If the density of dense PyC layer is lower than 1.6 g/cm³ the inner and outer PyC layers would densify so rapidly during irradiation that stresses would be built up which are higher than those for a fracture of the layers.

Fig. 2 shows the variation of post-irradiation density as a function of pre-irradiation density when different hydrocarbon gases are used. When the pre-irradiation density increases the post-irradiation also increases, implying that a densification takes place during irradiation in general. At higher temperatures than about 800° C and with neutron fluences above 2.0 x 10^{21} n/cm², the densification rate becomes nearly constant as shown in Fig. 2.



Fig. 2 shows the variation of post-irradiation density as a function of pre-irradiation density.

4. Change in directional length of PyC coating layer by neutron irradiation

As shown Fig. 3 [11] and Fig. 4 [12], PyC coating layers show different behaviors in directional length change with different hydrocarbon source gases; namely methane and propane. However, for both hydrocarbons, as the pre-irradiation density increases the length change parallel to the deposition direction remains negative but the length change perpendicular to the deposition direction becomes positive above the density of about 1.8 g/cm³. Also, it is to be noted that when the neutron fluence increases the length change also increases.



Fig. 3. Variation in lengths of PyC layer from methane as a function of pre-irradiation density (pr and pp mean parallel and perpendicular to the deposition direction, respectively).

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Fig. 4. Variation in lengths of PyC layer from propane as a function of pre-irradiation density (pr and pp mean parallel and perpendicular to the deposition direction, respectively).