Mechanical Properties and Structures of Pyrolytic Carbon Coating Layer in HTR Coated Particle Fuel

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1. Introduction

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.

While the fuel kernel is a source for a heat generation by a nuclear fission of fissile uranium, each of the four coating layers acts as a different role in view of retaining the generated fission products and the other interactions during an 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.

Although a recent review work [2] has contributed to an establishment of relationship between the material properties and QC measurements, the data on the mechanical properties and structural parameters of PyC coating layers remain still unclearly evaluated. A review work on dimensional changes of PyC by neutron irradiation was one of re-evaluative works recently attempted by the authors. [3] In this work, an attempt was made to analyze and re-evaluate the existing data of the experimental results of the mechanical properties, i.e., Young's modulus and fracture stress, in relation with the coating conditions, density and the BAF (Bacon Anisotropy Factor), an important structural parameter, of PyC coating layers obtained from various experiments performed in the early periods of the HTR coated particle development.

2. Young's modulus and fracture stress of PyC coating layer

A number of mechanical and physical properties such as elastic constants and the thermal conductivity are affected by density of PyC deposited by a fluidized bed CVD coating, which must be controlled in order that particles behave satisfactorily during irradiation. Density of PyC layer is usually controlled by the CVD deposition condition.

Mechanical properties of an as-deposited PyC layer, connected somehow with its density and other structural parameters, have been investigated extensively along with the works performed for the structural studies of its strength and elastic modulus. In early works carried out in the 1960's, most experiments for a PyC coating were concerned with methane as a source hydrocarbon gas, and the mechanical properties investigated with these experiments were based on a methane-derived PyC. [4, 5, 6] These measurements include the data for a low temperature- and high temperature-deposited PyC with various methane concentrations and temperatures ranging between 1673 – 2173 K and 2273 – 2673 K. Later, until the mid-1970's, the investigations on the mechanical properties were extended for source gases to propane, propylene and also a mixture of acetylene and propylene gases at low temperatures, which enabled one to collect data based on a propane- and a propylene-derived LTI PyC. Particularly, Kaae compared relationships between the structure and properties of different PyC's by taking the experimental data with methane from the work by Bokros and Price [5] and Price [6], and those with propane from his work.[7] For a acetylene-propylene gas mixture, he used unpublished data.[8]

2.1 Young's modulus

Fig.1 shows the variation of Young's modulus (solid symbols) as a function of PyC coating density, depending on the fraction of source gas and temperature. The values given in parentheses indicate the BAF values obtained for the given deposition conditions. It reveals that the modulus increases with the density of PyC coating layer for LTI-PyC, showing higher modulus ($20x10^3$ MPa < E < $50x10^3$ MPa) than HTI-PyC with lower fraction of source gas. (E < $20x10^3$ MPa) Although HTI-PyC could be obtained high density with low BAF (isotropic), the Young's moduli for HTI-PyC were low. LTI-PyC obtained using large fraction of propane shows comparable densities and modulus to those obtained with HTI-PyC in large fraction methane.

2.2 Fracture stress

Fig.2 shows the variation of fracture stress and BAF as a function of PyC coating density with various deposition conditions. The dependency of a fracture stress on the density seems unclear, as Fig. 2 shows: For LTI-PyC with methane, it has a value between 280 and 490 MPa, while it is between 175 and 210 MPa for HTI-PyC with comparable densities. When propane was used as a source gas, the LTI-PyC obtained at

temperatures below 1823 K showed a tendency of an increasing fracture stress together with modulus with an increasing density. This result would need some more detailed and systematic experiments for a re-evaluation.

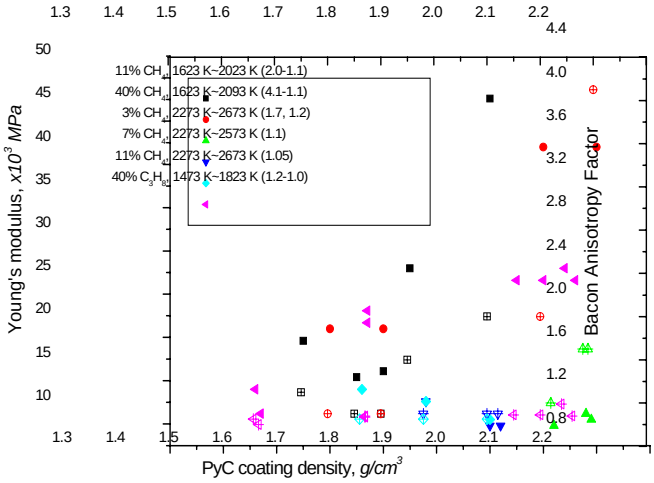

Fig. 1. Variation of Young's modulus and BAF as a function of PyC coating density with various deposition conditions (modulus : solid symbols, BAF : open-cross symbol)

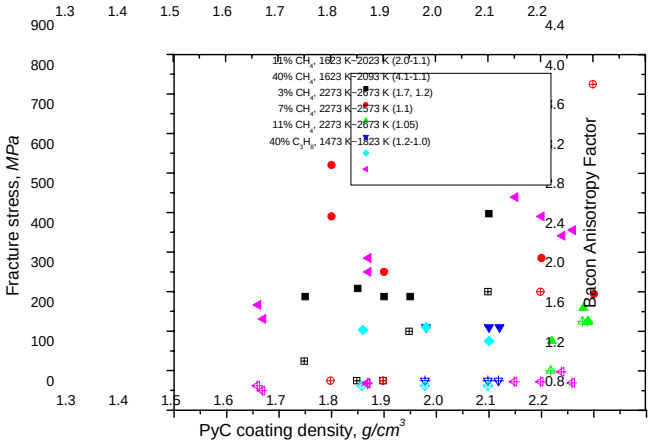

Fig. 2. Variation of fracture stress and BAF as a function of PyC coating density with various deposition conditions (fracture stress : solid symbols, BAF : open-cross symbol)

3. Conclusions

From the review and analysis in this work, the following important conclusions can be drawn.

- In the early developmental period, the mechanical properties of PyC were mostly measured by using PyC specimens derived from methane. However, the currently used PyC's are either acetylene-derived for the porous buffer layer or an acetylene/propylene mixture or propylene for the dense PyC layers for SiC-TRISO coated particle fuels.
- Even though a PyC deposited with different hydrocarbon gases show apparently similar characteristics, structural properties do not reveal the similar material properties. Material properties should be carefully selected for their use for the evaluation of obtained PyC layers.
- It can be suggested that the current PyC deposits used for manufacturing coated particle fuels be re-evaluated for their material properties and measurement data.

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