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A Novel Approach to the Porosity Evaluation of Pyrolytic Carbon Layers in TRISO-coated Fuel

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1. Introduction

The coating layers surrounding the fuel kernels in the spherical TRISO coated particles, consisting of pyrolytic carbon (PyC) and silicon carbide layers, act as a pressure vessel that retains fission product gases [1]. Accurate estimates of out-of-pile and in-pile material properties of the coated particles are critical to the development of robust and reliable fuel performance models [2]. Even though the various measuring techniques have applied to evaluate the properties of the coated layer in the TRISO coated particles, the better evaluation methods will be continuously requested to get advanced data. The sink float method and the dimension measurement apply to evaluate the density of the PyC layer in the TRISO coated particles. But these methods have some limitations like accuracy and difficulty of the sample preparation. Therefore, we try a new approach to get the porosity of the PyC layers with various pores in the sphere coated particles using the several reported equations, which was described the relation between the elastic moduli and the porosity. The elastic moduli could be obtained by the depth-sensing indentation method [3]. And then, using the measured elastic moduli, the porosity was calculated by selecting reliable one of the reported equations.

2. Experimental Procedure

The TRISO coating was performed on the ZrO_2 spheres as surrogates for UO_2 particles using C_2H_2/Ar, C_2H_2/C_3H_6/Ar, and CH_3SiCl_3/H_2/Ar, respectively, in a fluidized CVD coater. For measuring the elastic moduli, the polished cross-sections of the coated particles were prepared. The depth-sensing indentation was carried out with the nano-indentation device (NanoTest, Micro Materials Ltd.,) and a Berkovich diamond tip. The indentation was tested with the maximum penetration depth of diamond tip (about 1000 to 1500 nm) in the device, which would reflect the influence of pores to the utmost in pyrolytic carbon layer. Plots of load-depth curves, representative of the depth-sensing indentation data obtained from dense PyC (inner PyC) and porous PyC (buffer), are presented in Fig. 1. After indentation, a highly elastic recovery with no plastic deformation was demonstrated in both the dense and porous PyC materials, even at a penetration depth of ~ 1500 nm. The elastic moduli were derived from the initial portion of the slope in the unloading curve. Detailed explanation on the calculation of indentation elastic moduli was in elsewhere [4].

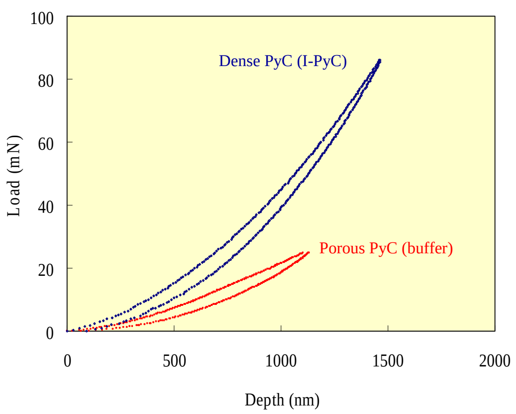

Fig. 1 Load-depth curves of Inner and buffer PyC layer obtained by depth-sensing indentation test.

3. Dependence of Elastic Moduli on Porosity

The dependence of Young's modulus, E, on porosity, P, has been studied for many materials. It has been proposed by many researchers that the relation between porosity and elastic moduli can be expressed as several empirical equations depended on pore shape, grain morphology and structural geometry [5]. In this work, the following polynomial equation of Coble and Kingery [6] was used on the basis of the boundary condition E = 0 at P = 1.

$$E = E_0\,(1 - 1.91P + 0.91P^2)$$

E and E_0 are the elastic moduli at the porosity = P and zero, respectively.

3. Results and Discussion

Fig. 2 shows distribution of the elastic moduli from one of TRISO particles with complete four components; Outer PyC / SiC / Inner PyC / Buffer. The indentation tests started from SiC to buffer layer. The corresponding microstructure is placed at the bottom side in Fig. 2. The indenting at the site of SiC layer makes the residual indentation impression with identical distance, which gives good information on the estimation of indenting point at the pyrolytic carbon layer. In this TRISO

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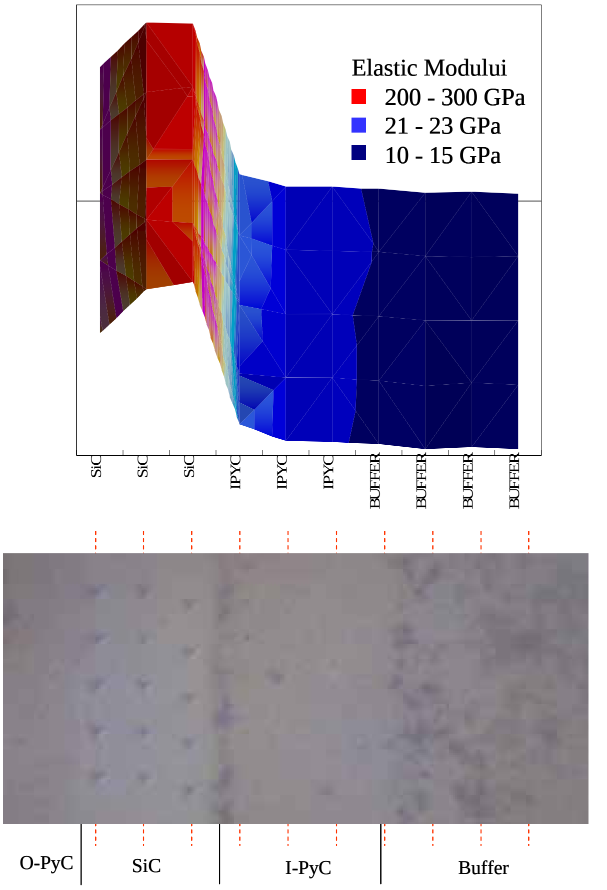

Fig. 2 Elastic moduli of SiC, I-PyC and buffer layer in TRISO particle corresponding to the microstructure.

particle, the elastic moduli of inner PyC and buffer layer exhibits about 22 GPa ± 1 and 12.5 GPa ± 1.5, respectively. Highly scattered data of elastic moduli are exhibited in near boundary region of SiC and PyC, because of mixture of SiC - PyC grains and pores with a high density. Fig. 3 shows the calculated porosity of inner and buffer PyC corresponding to the elastic moduli. The tendency of porosity was well agreed with the microstructural morphology as shown in Fig 2.

Diverse TRISO particles were fabricated by controlling input gas ratio and temperature, which provided several types of pyrolytic carbon layers with different porosities. The elastic moduli of the PyC layers in the range of 8.6 to 22.9 GPa with the porosities were obtained from the depth-sensing indentation tests. The calculated porosities of the corresponding PyC layers exhibited in the range of 43.7 to 4.5 %. The novel method introduced in this work has an advantage to obtain the porosity from the completed TRISO particle. In addition, uniformity of the pore distribution can be estimated at each layer of TRISO particle.

4. Conclusions

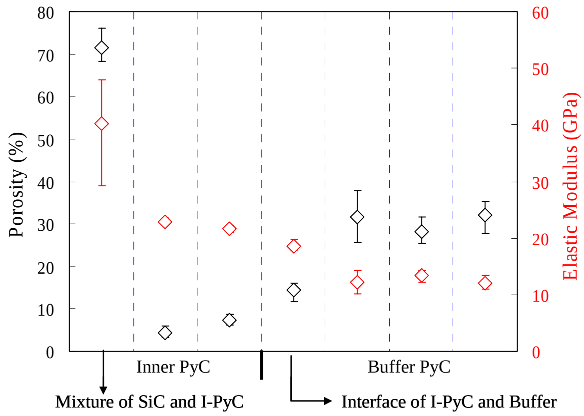

Fig. 3. Porosity calculated from the elastic moduli at the inner and buffer PyC.

A new approach was introduced to obtain the porosity of the PyC layers with various pores in the sphere coated particles. The relation between the elastic moduli and the porosity gave the effective data on the porosity of the completed TRISO particles using one of the reported equations. The porosity increase causes the reduction of the elastic moduli of the PyC layer. Tendency of the calculated porosity was well agreed with the microstructural morphology. Further studies will be characterized by comparing the porosity results measured by different techniques.

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