

Strength of chemical vapor deposited silicon carbide films using an internal pressurization test

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Silicon carbide coatings for tri-isotropic (TRISO)-coated fuel particles were fabricated using a chemical vapor deposition process at different deposition temperatures. An internal pressurization mechanical testing method, devised to measure the strength of the silicon carbide coatings, was investigated. An enhanced strength equation was derived using finite element analysis and verified by an experimental method utilizing a soda-lime glass standard specimen. The strength and Weibull modulus of the silicon carbide were measured using the internal pressurization technique. The effect of the microstructure and deposition temperature on the strength of the silicon carbide coating is discussed. Finally the reliability of silicon carbide coatings for tri-isotropic-coated fuel particles was discussed in terms of the coating strength with a correlation of the effective surface area and Weibull statistics.

Key words: Silicon carbide, Coating, Chemical vapor deposition, TRISO, Strength, Internal pressurization, Weibull statistics.

Introduction

Silicon carbide has good properties such as high strength, high temperature strength, low electrical conductivity, low thermal decay, low tritium permeability, and resistance to neutron irradiation [1, 2]. Recently advances of silicon carbide fiber reinforced silicon carbide composites show developments toward practical applications in nuclear power plants for fusion blankets and as functional materials [3, 4]. Chemically-deposited silicon carbide films have gained interest for utilization as a pressure vessel in tri-isotropic (TRISO)-coated fuel particles. TRISO-coated fuel particle consists of three pyrolytic carbon layers and one silicon carbide layer which retain the fission product gases in the kernel. When a TRISO-coated fuel particle is subjected to high temperature operation, the internal pressure of the particle is increased and a high hoop stress is concentrated on the silicon carbide layer [5]. Therefore the strength of the silicon carbide layer is one of the most important factors for the safer design of a fuel particle [6].

The failure modes of TRISO-coated fuel particle have been analyzed in a previous study and the strength of the silicon carbide layer was regarded as a primary physical property [6, 7]. Recently several studies on the strength of chemical vapor deposited silicon carbide films have been reported. Hong *et al.* suggested an internal pressurization and a crush test and reported the strength for silicon carbide films of cylindrical and hemispherical shell shapes in

terms of their effective surface areas [4]. A trilayer testing method was suggested by Kim *et al.* and the effect of the microstructure on the strength of silicon carbide was discussed [7]. The difficulties in the determination of the strength of silicon carbide layer in a TRISO-coated fuel particle still remains because of the small scale of the specimen geometry and ambiguity of loading configurations.

In this study, an internal pressurization test method to determine the strength of the silicon carbide film is suggested. The suggested method was based on the conventional bulge test [8, 9] and an accurate strength equation was derived using finite element analysis. Silicon carbide films were fabricated by a chemical vapor deposition process at different deposition temperatures [10, 11]. The strength of each specimen was evaluated with the suggested strength test method. Finally, the reliability of fabrication of each specimen is compared and the optimum microstructure for a TRISO-coated fuel particle is discussed.

Experimental Procedure

Materials

Silicon carbide coatings and soda-lime glass were used in this study. Four grades of silicon carbide coatings were fabricated onto graphite substrates. A conventional hot-wall type low pressure chemical vapor deposition technique was used to deposit the silicon carbide coatings. The source gas was methyltrichlorosilane, (MTS, CH_3SiCl_3), and the dilution gas was hydrogen. MTS and H_2 were mixed with a ratio of 5 : 1, and the total flow rate was fixed at 800 sccm. The pressure in the reaction chamber was maintained as 2.67 kPa, and the deposition time was 1 hour. Three deposition temperatures, 1200, 1300, and 1400 °C, were

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selected to fabricate silicon carbide coatings with different microstructures. The microstructures and phases of the silicon carbide coatings were characterized by scanning electron microscopy and X-ray diffractometry. For the suggested strength testing, as-deposited specimens were exposed to air in a furnace to eliminate the graphite substrate and obtain free-standing silicon carbide coatings. The temperature of the furnace was 700 °C and the duration time was 4 hour.

Soda-lime glass was used for the validation of the suggested strength test because its strength value is well-known and its nature of brittle fracture is similar to the failure of silicon carbide brittle coatings. Soda-lime glass was cut into 10 mm × 10 mm squares and a thickness of 0.18 mm. One side of soda-lime glass was abraded lightly with silicon carbide abrasive powder of grit 600, which induces uniformly distributed flaws on the surface of glass. Abraded soda-lime glass has a strength of 100 MPa which was reported elsewhere [12].

Test method

The internal pressurization strength test fixture was designed as a piston-like structure. The fixture was made with stainless steel with a 2 cm diameter hole and the diameter of the opposite hole was 4 mm in diameter. The silicon carbide coating and soda-lime glass were adhered onto the smaller hole with epoxy resin. Uncured polymethylmethacrylate (PMMA) liquid was filled into the hole as a pressure medium. The piston was pressed into the hole and induced a hydrostatic pressure inside the hole and specimen. The uniform pressure was applied to the bottom surface of the specimen. The specimen attached to the fixture was mounted on a universal loading machine and the piston was pressed down at 0.1 mm/minute crosshead speed until the specimens failed and a pressure drop occurred. The critical load for the failure was obtained and converted into a pressure value by dividing the critical load by the area of the piston. A detailed analysis of the test method is described in the following section.

Analysis of the test method

If a flat plate specimen is internally pressurized by a pressure p induced by the hydrostatic compression of a viscous fluid, assuming a linear, homogeneous, and isotropic material, the hoop stress is found from elastic of plate theory to be a function of the radial distance from the center of a disc specimen a :

$$\sigma = \frac{3}{4} p \left(\frac{a}{t} \right)^2 \quad (1)$$

where t is the thickness of the specimen and a is distance from the center. This relation is derived under the boundary condition of clamped edges. The fracture stress, which corresponds to the maximum tensile hoop stress when the fracture occurs, is given as the stress at the outer ring edge. However, the maximum stress at the outer ring edge

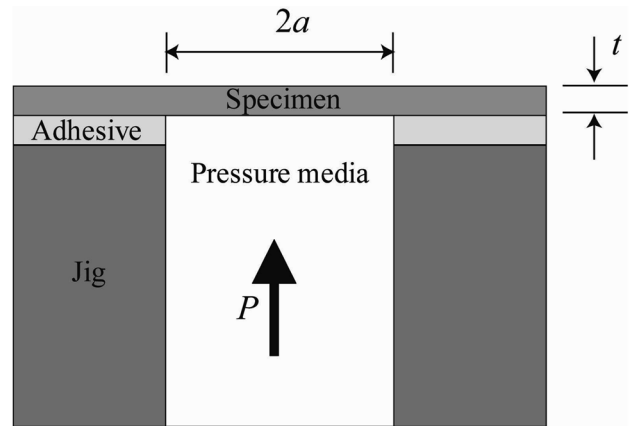


Fig. 1. Schematic drawing of the internal pressurization test method. The coating is adhered on the fixture with epoxy adhesive. The inside of the fixture is filled with uncured PMMA liquid and pressurized by the piston.

does not match exactly the stress which is given by the analytical equation because an adhesive interlayer is inserted between the fixture and plate. This ambiguous boundary condition induces a change of the stress profile along the radial distance. In order to obtain the precise stress profile and the maximum tensile hoop stress, finite element analysis was used to simulate the test configuration. In the analysis, an axi-symmetric model with four node bilinear axi-symmetric quadrilateral elements is used as the configuration and the maximum mesh size is less than 1 μ m. A detailed schematic diagram of the test fixture is shown in Fig. 1. All materials used in the finite element analysis are assumed to be linear elastic and the pressure is described as a uniform surface pressure at the bottom of the plate. As a result, the exact maximum hoop tensile stress is obtained and compared with the analytical solution. A precise fracture stress equation is derived by introducing the radius and stress compensation parameter according to the hoop stress of the finite element analysis:

$$\sigma = \frac{3}{4} p \left(\frac{a-\alpha}{t} \right) \beta \quad (2)$$

where α is 40 and β is 0.9. These parameters were determined by a regression fit to the finite element analysis data. The maximum stresses with different coating thicknesses, moduli and adhesive thicknesses are shown as data points and the suggested equation is shown as a solid line in Fig. 2.

It is well-known that brittle solids show a catastrophic failure with stress concentrations and their strengths depend on the size and surface condition of the specimens. A statistical distribution is classically applied to brittle solids, Weibull statistics, combined with fracture mechanics, is typically used to describe the failure in this study. Weibull statistics, otherwise described as the weakest link theory, describe the failure of a material as the propagation of a single flaw, like a chain failing through the breaking of a

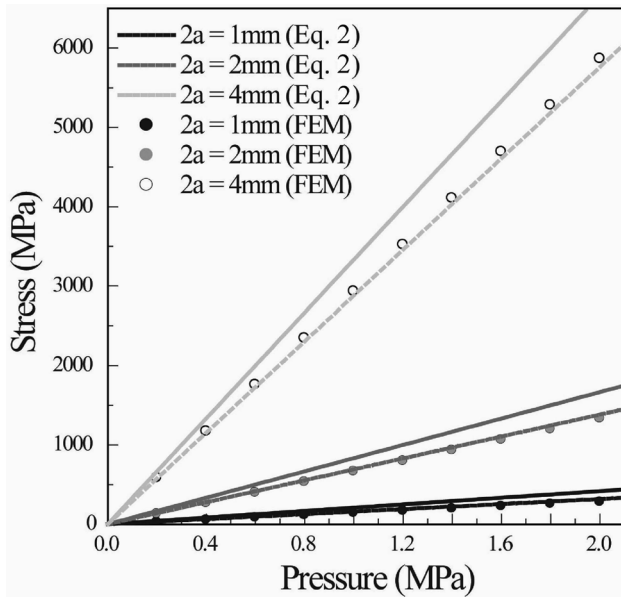


Fig. 2. Finite element analysis results (data points) and analytical and suggested equations (dashed and solid lines). This solid curve are for fits to Eq. (2).

single weak link. The most serious flaw is not necessarily the largest flaw in that other factors contributing to failure need to be considered such as the orientation with respect to stress. For the case of a critical flaw in a coating plate, the Weibull distribution is given by:

$$P = 1 - \exp\left[-S_E \left(\frac{\sigma_{max}}{\sigma_0}\right)^m\right] \quad (3)$$

where P is the cumulative probability of failure, σ_{max} is the maximum stress, m is the Weibull modulus, and σ_0 is a scale parameter. By taking the logarithm twice, Eq. (3) can be rewritten in a linear form:

$$\ln\ln\left(\frac{1}{1-P}\right) = m \ln\sigma_{max} + \ln\left(\frac{S_E}{\sigma_0^m}\right) \quad (4)$$

The Weibull modulus can be obtained from the slope in Eq. (4). As the true value of P_i for each σ_i is not known, a prescribed probability estimator has to be used as the value of P_i . Among probability estimators, it is shown that the probability estimator of Eq. (5) gives a conservative estimation, from an engineering point-of-view, it should be the best choice in reliability predictions:

$$P_i = \frac{i}{N+1} \quad (5)$$

where P_i is the probability of failure for the i th-ranked stress datum and N is the sample size. Finally the strength and Weibull modulus can be converted into the estimated strength of TRISO-coated fuel particles as:

$$\sigma_2 = \left(\frac{S_{E2}}{S_{E1}}\right)^{-1/m} \cdot \sigma_1 \quad (6)$$

where subscript 1 is the original value and 2 is the converted value. The effective surface area of TRISO-coated fuel particles was determined from the surface area of the silicon carbide coating and the effective surface of the pressurized coating layer is obtained from the stress profile of finite element analysis. The position of 90% of maximum stress is regarded as a boundary of the effective surface area. The reliability of a TRISO-coated fuel particle is analyzed with this converted strength value [13].

Results and Discussion

Microstructure

Silicon carbide coatings fabricated by chemical vapor deposition at 1200, 1300, and 1400 °C were used in this study. A scanning electron microscope was used to observe the microstructure of the silicon carbide coatings. Top surface views of each silicon carbide coating are shown in the right side of Fig. 3. The silicon carbide coating deposited at 1200 °C shows an isotropic and dome-top shaped microstructure. A faceted grain structure is obvious in the 1400 °C specimen and a mixed microstructure is shown in the silicon carbide coating fabricated at 1300 °C. The microstructure tends to be isotropic at a low deposition temperature and faceted at high deposition temperature. X-ray diffractions pattern for each specimen are shown in the left side of Fig. 3. The silicon carbide coating used in this study was primarily the beta form and 3C cubic phase. The preferred orientation of the silicon carbide coating deposited at 1200, and 1300 °C was the [111] direction and only the [220] direction was observed in the specimen fabricated at 1400 °C. These differences of microstructure and preferred orientation are due to the deposition mechanism which is reported elsewhere [10, 11]. A side view of the specimen deposited at 1400 °C showed a columnar microstructure and the other specimens showed isotropic microstructures [10, 11]. The strengths of these specimens were evaluated with the proposed internal pressurization method.

Validation of suggested equation

Prior to the strength test of silicon carbide coatings, the proposed method was validated with standard specimen. To confirm the validation of the proposed equation, a soda-lime glass with a strength of 100 MPa was selected as a standard specimen because of its brittle fracture behavior. In Fig. 4, a Weibull diagram of strengths for soda-lime glass specimens obtained from the proposed testing method is shown. According to the results, the proposed internal pressurization method was verified successfully. In the Weibull diagram, a regression fit to data points yields a Weibull modulus $m = 6.88$ which is reasonable for the brittle solids. This Weibull modulus is typical for the soda-lime glass which was lightly abraded with silicon carbide abrasive powder of grit 600. Care must be taken if different materials are used for

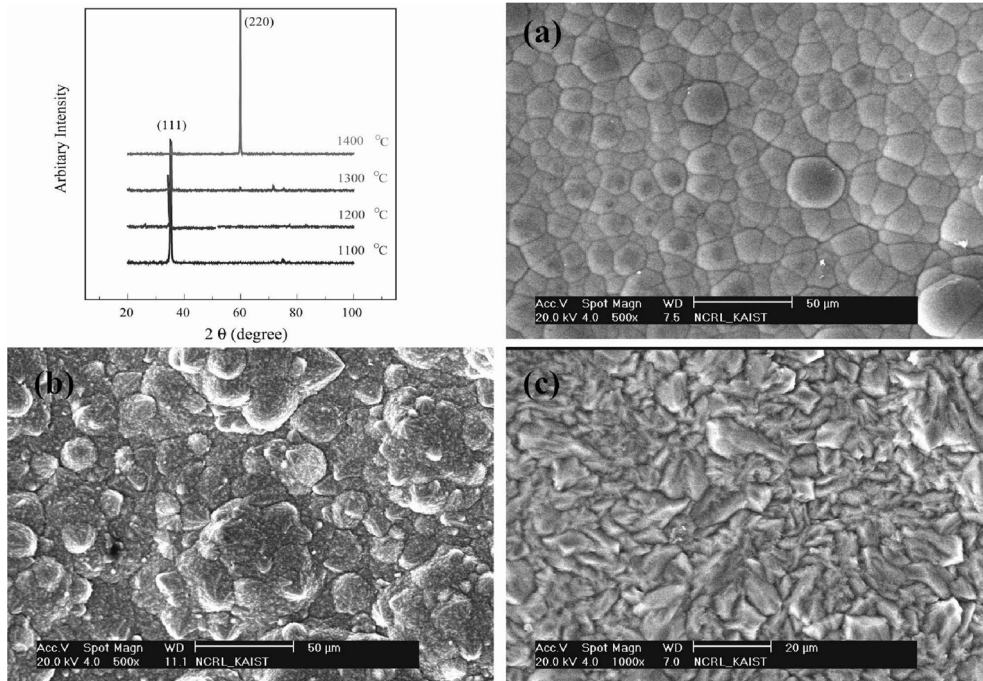


Fig. 3. Microstructures of the deposited silicon carbides (a), (b), and (c) at 1200, 1300, and 1400 °C, respectively, observed by a scanning electron microscope (right side), and each X-ray diffraction pattern (left side).

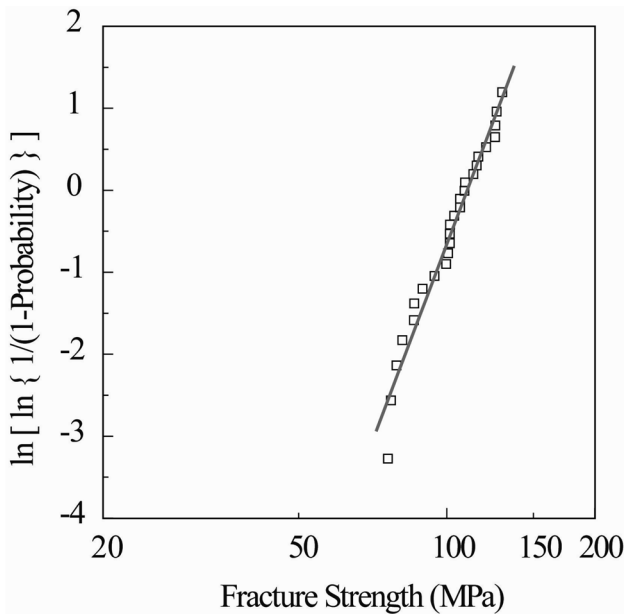


Fig. 4. Experimental data showing the strength of soda-lime glass as a form of Weibull statistics. The solid curve is calculated from Eq. (4).

the testing fixture and adhesive, as the equation is derived from finite element analysis with a fixed modulus of the fixture and the adhesive. In situ observation of the failure of soda-lime glass reveals that the crack initiates from the edge of the specimen and propagates along the circle-shaped edge. This fact implies that the maximum stress occurs at the edge of the specimen and the failure stress analysis is valid.

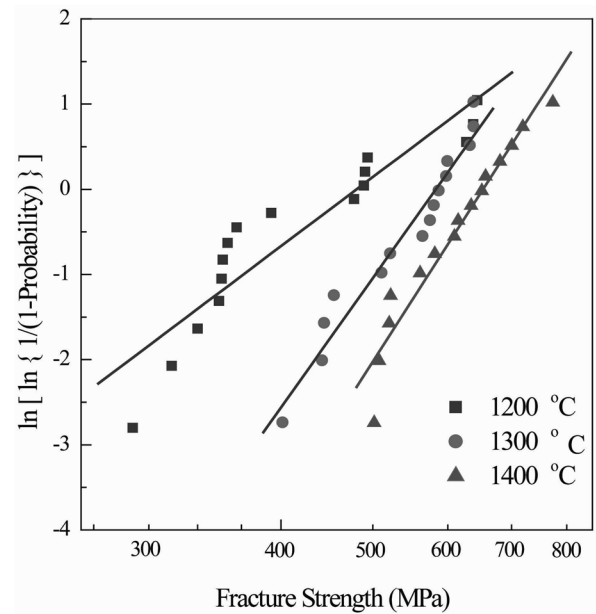


Fig. 5. Experimental data showing the strength of silicon carbide coatings deposited at 1200, 1300 and 1400 °C. The solid curves are calculated from Eq. (4).

Strength and Weibull analysis

In this study, we applied an internal pressurization test method to compare the strength of three types of silicon carbide coatings. Strength values are displayed for each specimen in Fig. 5. as a form of Weibull statistics. For this statistical analysis, fifteen specimens were tested. In the case of the silicon carbide coating deposited at 1200 °C, the strength of the specimen was 484 MPa and

the Weibull modulus was 3.69. In a previous study on the strength of chemical vapor deposited silicon carbide, the strength value was over 500 MPa and Weibull modulus was over 7 [7]. In comparison with this previous result, our strength value is low and the Weibull modulus is quite low. From the Weibull modulus we can estimate that the reliability of this silicon carbide coating is not sufficient for safe coating of TRISO fuel particles. In the case of the silicon carbide coating fabricated at 1300 °C, the strength value was 584 MPa and Weibull modulus was 6.92. The strength value was improved and Weibull modulus was also increased. According to the strength and Weibull modulus, a silicon carbide coating deposited at 1300 °C is acceptable for TRISO-coated fuel particles. The strength of 654 MPa and Weibull modulus 7.66 was obtained for the silicon carbide coating deposited at 1400 °C. In general, the reliability of a silicon carbide coating is largely improved with an increase in the deposition temperature.

The difference of strength value of each specimen may be due to the microstructure of coatings. With a low deposition temperature, the surface of the coating layer is rough and the microstructure was not fully developed. The low strength and Weibull modulus can be explained with these imperfections in the microstructure. In the case of silicon carbide coatings deposited at 1300 and 1400 °C, the strength and Weibull modulus have acceptable values to make mechanically-reliable TRISO-coated fuel particles. According to the strength results, a higher deposition temperature is desirable to improve the strength and Weibull modulus. In an early study concerning the failure probability of TRISO-coated fuel particles, the Weibull modulus was found to be an important factor to reduce the failure probability of TRISO-coated fuel particles [5]. High Weibull modulus value is necessary for safer design of TRISO-coated fuel particle.

Prior to discussing the reliability of silicon carbide coatings, their effective surface areas should be considered. For TRISO-coated fuel particles, a uniform pressure is applied on the entire sphere surface and its surface area is around 5.06 mm² with a 0.635 mm radius. In the internal pressurization test method, the effective surface area is around 0.74 mm². The strength obtained from this study needs to be converted to a value for TRISO-coated fuel particles. Using Eq. 6, we can calculate the converted strength for the three silicon carbide coatings and the maximum converted strength is 510 MPa (for the specimen deposited at 1400 °C) which is slightly lower than the value obtained. The converted strength value is still sufficient to retain the kernel overpressure during the fusion reaction. From a structural point-of-view, the silicon carbide coatings deposited at 1300 and 1400 °C are good candidate materials.

Conclusions

Silicon carbide coatings for TRISO-coated fuel particles were fabricated by a chemical vapor deposition method. In order to evaluate the strength of the silicon carbide coatings, an internal pressurization test was suggested and an equation for the calculation of strength was obtained by a regression fit to the data from a finite element analysis. The silicon carbide coating deposited at a high temperature showed a higher strength and Weibull modulus. An effective surface area concept was adopted to determine the reliability of the silicon carbide coatings. It was concluded that silicon carbide coatings deposited at high temperature are good candidates for the safe design of TRISO-coated fuel particles.

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