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Helium Gas Permeability Evaluation of Triplex Nuclear Fuel Cladding

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1. Introduction

Stability of a nuclear power is of increasing concern after 2011 Fukushima accident. The direction of technology development has changed from the economic feasibility improvement of the nuclear fuel under a normal operation condition to the accident tolerance reinforcement of the nuclear fuel cladding under a severe accident. The accident tolerance enhanced nuclear fuel system is needed to satisfy two parts. First, the performance has to be retained compared to the existing UO_2 nuclear fuel and zircaloy cladding system under the normal operation condition. Second, under the severe accident condition, the high temperature structural integrity has to be kept and the generation rate of hydrogen has to be reduced largely. Multi-layered SiC composite fuel cladding, which consists of monolith inner/outer layer and intermediate SiC_f/SiC composite layer, is one of candidates for a replacement for the zirconium alloy cladding owing to the superior high temperature strength and low hydrogen generation under severe accident conditions. The precise performance test of SiC composite cladding under a normal operation and severe accident condition is required in order to apply the SiC composites cladding to an accident tolerance fuel cladding. Gas permeability test for nuclear fuel cladding is one of the important performance tests. Gas hermeticity of SiC composite cladding in an as-fabraicated condition as well as a normal and a severe accident condition could be obtained. A few studies have been reported in gas permeability of SiC/SiC composite tube, however, has not been studied well in a SiC composite cladding[1,2].

Gas hermeticity which is represented in leak rate means gas that flows towards inside of a vacuum system for time unit. Helium is used as the trace gas for detecting small leaks. Helium is nontoxic, does not react with other elements and can even penetrate through the smallest place due to its very small size atoms. Helium is exist only in very small quantities in the air and so air does not substantially disturb the measurements. Gas permeability is the intrinsic property of own materials, shows the property that pass through inside a material under pressure difference. In a fusion energy system with SiC/SiC composites, helium gas has been considered as a coolant. Therefore, it is an important issue to evaluate helium gas permeability of SiC/SiC composite.

In this study, gas leak test of CVD-SiC tube and SiC composites tube(Triplex tube, inner SiC/SiC composite/outer SiC) fabricated by CVD and CVI was conducted. The equipment for gas leak test which enable to evaluate helium gas leak rate on tube specimen under the condition of constant pressure was developed using helium leak detector. Gas leakage part was observed by microstructure investigation of the specimen. Helium gas permeability was calculated from helium gas leak test. Gas permeability of triplex tube after the hoop strength test and thermal shock experiment which are designed to simulate the condition of a normal operation and a severe accident was evaluated.

2. Methods and Results

Gas leak test is commonly used experiment which is evaluated a gas hermeticity of the joining parts using helium gas detector. Helium leak rate was measured with the help of Variant helium leak detector (VS PRO2). Maximum measurement leak rate is 9.9 x 10^{-4} atm·cc/s, minimum measurement leak rate is 1 x 10^{-10} atm·cc/s. Gas leak test equipment is composed of upper chamber, down chamber, middle part, and vacuum system. The ends of SiC tube were closed with stainless steel plug. Epoxy resin was used to bond the tube and stainless steel plug. The middle part(stainless steel) plate has a hole at the center to facilitate vacuum application in the tube. The test was conducted at room temperature and constant upper chamber pressure. The equipment has been designed to have a inner chamber pressure value from 0.1 atm to 1 atm. Gas permeability of specimen could be calculated from the gas leak test by the standardization of equipment and specimen. Unit of leak rate shown by helium leak detector is mbar·l/s, means the effective volume of helium gas coming through inside the detector for time unit. From the equation of ideal gas raw, leak rate(l) could be shown below equation.

$$l = \frac{P_{down} V_{down\,chamber}}{t}$$

P_{down} is the pressure of down chamber in the gas leak test equipment, $V_{downchamber}$ is the volume of down chamber in the equipment. In the gas leak test, helium gas permeability(K_{He}) could be below equation under the condition of constant upper chamber pressure(P_{upper}).

$$K_{He} = \frac{P_{down}}{P_{upper}} \frac{D}{A} S_{eff}$$

$$= \frac{P_{down}}{P_{upper}} \frac{D}{A} \frac{V_{downchamber}}{t}$$

$$= \frac{D}{A} \frac{1}{P_{upper}} l$$

D is the thickness of the specimen, A is the area of the specimen, S_{eff} is the effective pumping speed of the equipment. The tube specimen used for the leak rate experiments are CVD-SiC tube and triplex tube, which has a length of 30 mm, inner diameter of 10 mm. The effective length of specimen for leak rate was 20 mm because of bonding epoxy.

Helium gas leak test of CVD SiC tube and triplex tube was conducted. CVD tube has a thickness of 300 μm, 750 μm. Gas leak rate results were shown in Table 1. The condition of the pressure of upper chamber was 0.1 and 1 atm. In the gas leak test results of 750 μm CVD-SiC tube, any gas leakage was not measured in 0.1 and 1 atm upper chamber pressure condition. In the gas leak test results of 300 μm CVD-SiC tube, 2.0 x 10^{-9} atm·cc/s for 0.1 atm, 2.5 x 10^{-9} atm·cc/s was measured. In the reported gas leak rate results of CVD-SiC coating layer for Khalifa et al., helium gas leak rate of SiC coating layer which has a thickness of 140 μm shows about 1 x 10^{-9} atm·cc/s. It was observed that helium gas leak rate value according to the thickness of specimen showed similar value. Helium gas permeability of CVD-SiC tube material was calculated. In the 300 μm CVD-SiC tube, helium gas permeability is 9.4 x 10^{-15} m^2/s for 0.1 atm, 1.2 x 10^{-15} m^2/s for 1 atm. These results is similar value with helium gas permeability constant of common ceramic (10^{-12} ~ 10^{-15} m^2/s).

Table 1. Helium gas leak rate results

Condition	specimen	0.1 atm (atm·cc/s)	1 atm (atm·cc/s)
-	CVD SiC (750 μm)	0	< 0.1 x 10^{-9}
-	CVD SiC (300 μm)	2.0 x 10^{-9}	2.5 x 10^{-9}
Triplex tube	Triplex	0	< 0.1 x 10^{-9}
After hoop test	Triplex	7.1 x 10^{-5}	> 9.9 x 10^{-4}
After thermal shock	Triplex	3.3 x 10^{-4}	> 9.9 x 10^{-4}

In the gas leak test results of triplex tube under the condition of the upper chamber pressure 0.1 atm and 1 atm, any gas leakage was not measured. In the triplex tube, It is thought that two layers of inner SiC and outer SiC have sufficiently blocked helium gas penetration.

Helium gas leak test of triplex tube after hoop strength test and thermal shock test was conducted. Hoop strength test simulates inner pressure inside fuel cladding induced by fission product under a normal operation condition. Thermal shock test simulates the condition after a severe accident. Helium gas leak test results were shown in Table 1. Hoop strength test was conducted until initiating the crack in inner SiC layer of triplex tube[3]. Crack initiation load on inner SiC layer was measured by acoustic emission. From the hoop strength test, crack initiation in inner SiC layer occurs at 280 N. In thermal shock test, triplex tube was quenched from 1200 °C. The temperature of 1200 °C was decided because ECCS(Emergency Core Cooling System) operates at 1200 °C in an accident. The microstructure of the specimen after hoop strength test and thermal shock test was shown in Fig. 1. Crack of inner SiC layer was well observed in all specimens.

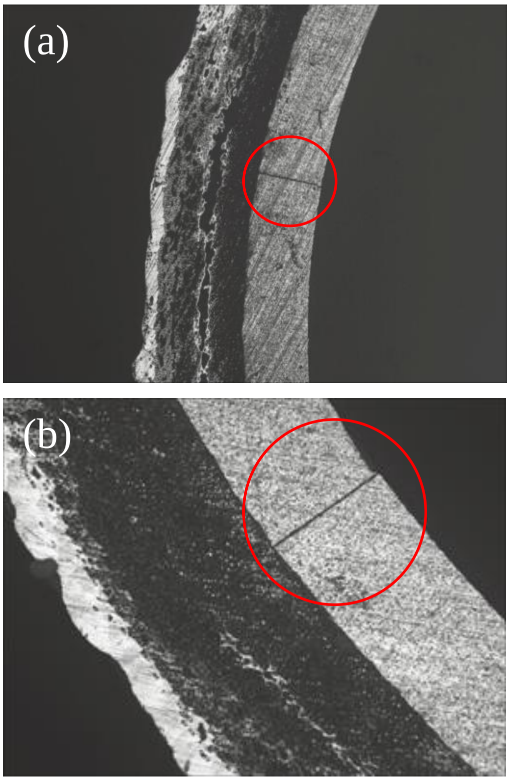

Fig. 1. Micrographs of tripled specimen after (a) hoop strength test (b) thermal shock test

In the gas leak rate results of the specimen after hoop strength, 7.1 x 10^{-5} atm·cc/s for 0.1 atm, above 9.9 x 10^{-4} atm·cc/s for 1 atm. These results mean that inner SiC layer falls in its role as gas hermeticity layer and only thin outer SiC layer performs a role as gas hermeticity layer. In the gas leak rate results of the specimen after thermal shock, 3.3 x 10^{-4} atm·cc/s for 0.1 atm, above 9.9 x 10^{-4} atm·cc/s for 1 atm. These results are similar with the results of the specimen after hoop strength test. At the results in the condition of 0.1 atm, there are a little difference of gas leak rate because outer SiC layers of each triplex sample has thickness inhomogeneity.

From the results, it is thought that helium gas hermeticity of triplex structure under a normal operating condition and a severe accident condition was not achieved. It is thought to need a duplex structure, which is composed of outer SiC layer and inner SiC_f/SiC composite, in order to prevent helium gas leak through fuel cladding.

3. Summary

Gas permeability evaluation method of triplex fuel cladding suggested by accident tolerance fuel cladding was studied. Helium gas leak test equipment was developed and helium gas permeability was calculated. Helium gas leak rate of CVD SiC tube, as-fabricated triplex tube and triplex after hoop strength test and thermal shock test was measured. In CVD SiC tube and as-fabricated triplex tube, any helium gas leakage was not detected. In triplex tube after hoop strength test and thermal shock test, a few leakages were detected because of cracks in inner SiC layer. Therefore, maintaining the helium gas hermeticity of triplex tube is thought to be difficult.

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