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Impregnation of the Polycarbosilane Preceramic Polymer into SiC Woven Fabric

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1. Introduction

Commercial nuclear fuel cladding tubes are currently made from various zirconium alloys. These alloys have been used because they show sufficient strength at normal operating conditions and have fairly low neutron capture cross sections. However, after the Fukushima accident, it was recognized that a hydrogen-related explosion is one of the major concerns of reactor safety during high-temperature corrosion of zirconium alloys. High interest has been focused on the stability of nuclear plants during accidents. It is therefore necessary to develop cladding tubes that have enhanced the stability in accidents

For existing zirconium alloys, oxidation occurs accompanied by a rapid corrosion when they reach 700–1200℃. Then, their strength abruptly degrades, generating large amounts of hydrogen. On the other hand, SiC has advantages such as stability in severe environments, lack of reaction with zirconium alloys up to 1500˚C, high thermal conductivity, low neutron absorption, radiation stability, and compatibility. SiC do not generate hydrogen since there is no oxidation. Recently, cladding tubes with a metal-ceramic duplex structure are being studied [1,2]. The new tubes form composite ceramic layers on the zirconium alloy cladding tubes to enhance their stability during all accidents, as well as to prevent them from generating hydrogen gas under severe accidents (see Fig. 1). The new tubes can be made in the following two stages: first, filament winding of SiC fibers on the zirconium alloy cladding tubes, and second, impregnating the polymers between the SiC fibers and the zirconium tube, and in the empty space in the filament wound SiC composite [3, 4]. This study aims at finding the optimal process conditions for the impregnation of polymers using a sol-gel method.

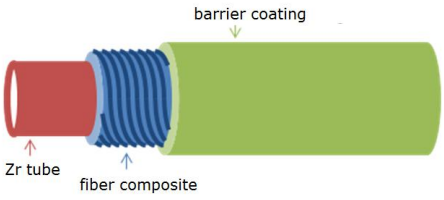


Fig. 1. Schematic illustration of metal-ceramic hybrid fuel cladding tubes.

2. Methods and Results

2.1 Methods

Preceramic polymer (PCP) with Si-C-O structures (ToBeMTech, Korea) was used to fill an open space within the fiber-wound layer. In the experiment, a two-dimensional fiber fabric of Tyranno-Lox M (Ube industry, twill weaved, Japan) was used for the process development. As sample for impregnation was prepared by wrapping the woven fabric on a zirconium tube, as shown in fig. 2. The prepared sample was dipped in a PCP-dissolved *n*-hexane solution. Polymer impregnation was performed under a low vacuum of 60–70 kPa. The immersed fabric was dried and cured in air or in a vacuum at 600℃.

Fig. 2. Model samples of the metal-ceramic hybrid cladding tube, used for impregnation

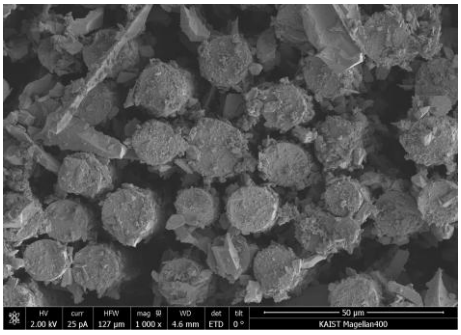
To optimize the conditions for the impregnation of the polymers, experiments were performed by varying the conditions, such as the kinds of PCP powder, the concentration of the solution, atmosphere (a vacuum, or not) drying, and the curing temperature. Furthermore, SiC nano powder was added, or the filling times were increased. The specimens obtained in this study were investigated and analyzed using a scanning electron microscope.

2.2 Polymer Impregnation

Fig. 3 shows images to compare the effect of curing atmosphere. Fig. 3(a) shows the specimen cured in a

vacuum. It was observed that the PCP was impregnated into the SiC fiber-wound layer. However, the density of impregnated polymer was low since the polymer was not cross-linked. On the contrary, in the case of curing in air (Fig. 3(b)), the density was increased as the cross-linking occurred.

(a)



(b)

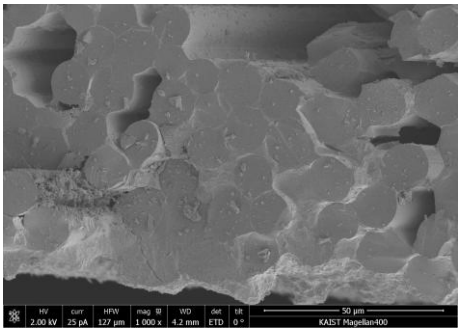


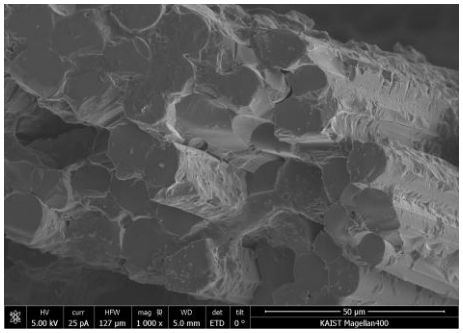
Fig 3. SEM images of SiC fabrics cured in (a) a vacuum and (b) air, respectively.

Fig. 4 shows the cross-sectional images of impregnated samples with different repeated times of impregnation and curing. Both Fig. 4(a) and (b) show a high rate of impregnation; however, it is revealed that the impregnation rate of samples that went through five cycles of impregnation and curing (Fig. 4(b) is higher than that of samples went through three repeated cycles (Fig. 4(a).

3. Conclusions

A preceramic Si-C-O polymer was impregnated into SiC-fiber fabrics. When cured in air, the density of the polymer was increased with the cross-linking of the polymer. In addition, the rate of impregnation was increased as the cycles of the impregnation and curing were repeated.

(a)



(b)

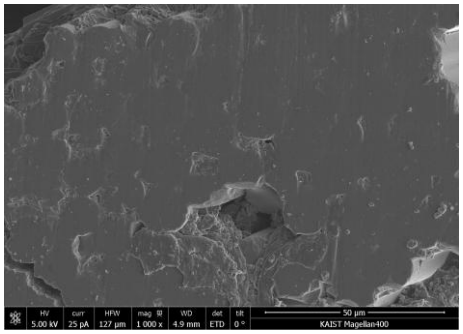


Fig. 4. SEM images of SiC fabrics (a) after three cycles of impregnation and curing, and (b) five repeated cycles

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