Integrity Assessment of SiC Oxidation at High Temperatures

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1. Introduction

In high burn-up PWR reactors, zirconium-based alloys are widely used as cladding material for fuel rods. This is because Zr alloys have a low neutron absorption cross-section, good corrosion resistance in various operating conditions, and satisfying mechanical properties. However, Zircalloy fuel cladding can cause accidents through oxidation and corrosion at high temperatures. Thus we need to discover several means to improve its oxidation and corrosion resistance. SiC is known to be one of a few materials with strong properties including resistance to corrosive and oxidative environments. It is an interesting method of placing a ceramic protective coating on the outer surface of the fuel cladding. If we can make a metal cladding with SiC composites as a protective layer, the use of the cladding can be very broad and diverse. Inherently safe nuclear fuels can be made possible that can withstand severe accident conditions. To date, no studies have discussed using to SiC composites to coat metal cladding. If we can make a metal cladding containing SiC composites, the SiC coating can act as a protective layer, allowing us to obtain an inherently safe nuclear fuel that can withstand severe accidents. In addition, we can expect positive economic effects in the nuclear industry.

2. Methods and Results

2.1 Specimen

Specimens (Fig. 1c) were obtained by dispersing the Tyranno SA SiC fiber (Fig. 1a) into a toluene-based solution and contacting 20% (weight ratio) of the associated Hydrido polycarbosilane (HPCS) (Fig. 1b).

Fig.1 (a) Tyranno SA SiC fiber, (b) 20% HPCS +80% toluene, (c) The final specimen

The SiC fiber was spooled many times and its dip coating placed into a 20% HPCS solution. After the dip coating dried for an hour at room temperature, it was cured for 2 hours in air at 273℃. It was then treated at 700℃ in a nitrogen atmosphere to eliminate hydrides from the HPCS. Figure 2 shows the procedure for making specimens.

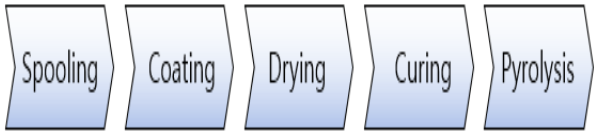

Fig. 2: Specimen-making procedure

2.2 Apparatus

To find oxidation and corrosion data, we used two pieces of experimental equipment: a TGA (Fig. 3a) and a tube furnace (Fig. 3b). Because I could not find any references to oxidation data on SiC fibers coated with HPCS solutions, we could only compare the results of the two experiments.

A TGA device can continuously measure weight changes. A tube furnace can only measure weight changes before, between, and after experiments.

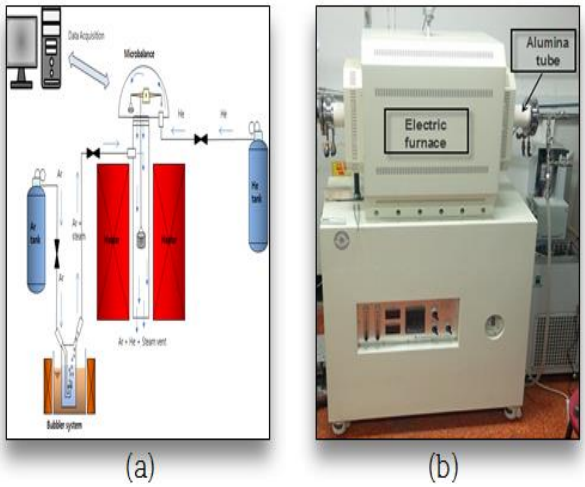

Fig. 3 (a) Thermo-gravimetric analysis (TGA) , (b) Tube furnace apparatuses

3 Results

Oxidation and corrosion experiments were performed in which the specimens were heated at 1200℃ in an air atmosphere (0.1Mpa) and steam for 20 hours. Figure 4

shows a SEM image of a SiC fiber dispersed into a toluene-based solution and contacting 20% of the HPCS. The cylindrical shape is the SiC fiber, and the HPCS coating is seen between the SiC fibers.

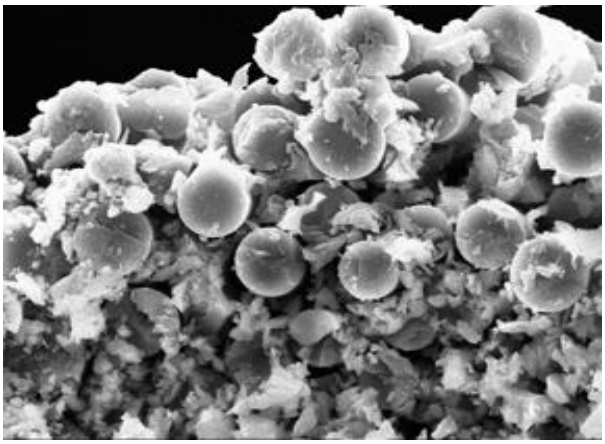

Fig. 4: SEM micrograph of the SiC fiber dispersed into a toluene-based solution and contacting 20% of the HPCS after pyrolysis

Figure 5 shows how specimens gained weight at 1200℃ in an air atmosphere (0.1Mpa) and steam for 20 hours. At the point of final weight gain in the air experiment, the TGA weight gain value was higher than the tube furnace's value. One reason for this is differences in air conditions between the TGA and tube furnace. In the TGA experiment, air was supported by air pumps, but the air in the tube furnace experiment was regular air. In the steam experiment, the final TGA weight gain was also higher than in the tube furnace. In the TGA, a little argon gas was injected in order to keep the micro balance from being damaged bt high temperatures during the long experimental time. At first, both the TGA and tube furnace weight gains decreased for 2 hours (steam) and 3 hours (air). In my opinion, this decrease range was caused by damage to the HPCS coating at high temperature (1200℃). After decreasing, the weight gain began to slowly increase, was higher for the air than for the steam.

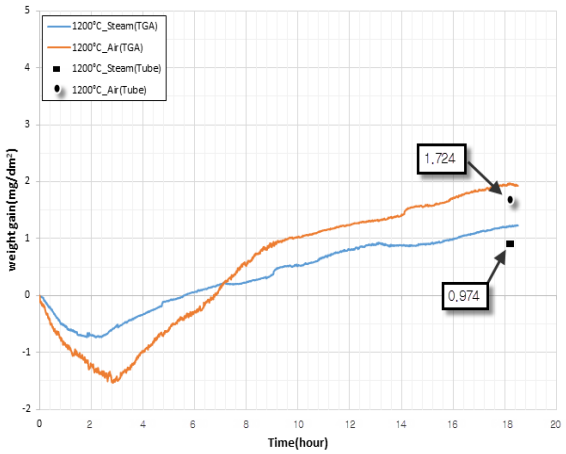

Fig. 5: Experimental results of the weight gain of specimens at 1200℃ in atmosphere (0.1Mpa) and steam for 20 hours

3. Conclusion

The oxidation experiment confirmed that SiC has good oxidation and corrosion resistance. Although there are differences in weight gain between TGA and the tube furnace, the weight gain for 20 hours was less than 2mg/dm^2, and the oxidation did not proceed when the HPCS coating was destroyed. The specimens also were not externally damaged at high temperatures for 20 hours. Even the SiC fiber was undamaged.

Oxidation experiments show that, when specimens are heated to 1200℃ in air for 20 hours, 1.9 mg/dm^2 is oxidized, while 1.2 mg/dm^2 is oxidized in steam. A SiC coating will improve the oxidation resistance of the cladding materils.

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