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Hydrothermal corrosion of silicon carbide joints without radiation☆

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ABSTRACT

Hydrothermal corrosion of four types of the silicon carbide (SiC) to SiC plate joints were investigated under pressurized water reactor and boiling water reactor relevant chemical conditions without irradiation. The joints were formed by metal diffusion bonding using molybdenum or titanium interlayer, reaction sintering using Ti–Si–C system, and SiC nanopowder sintering. Most of the joints withstood the corrosion tests for five weeks. The recession of the SiC substrates was limited. Based on the recession of the bonding layers, it was concluded that all the joints except for the molybdenum diffusion bond are promising under the reducing environments without radiation. The SiC nanopowder sintered joint was the most corrosion tolerant under the oxidizing environment among the four joints.

1. Introduction

Silicon carbide (SiC) fiber reinforced SiC matrix composites are considered as a primary candidate of the accident tolerant fuel cladding of light water reactors (LWRs) because of the outstanding irradiation and oxidation resistance, and low neutron absorption cross section [1,2]. For the successful development of the SiC based fuel cladding, joining the cladding to the end-plug is recognized as one of the technological hurdles [1,2]. One of the processing difficulties regarding the end-cap joining for a long cladding with thin wall thickness is a limitation in the applied stress which can be employed during the joining. Moreover, the functional requirements of the joint are (1) gas-tightness to retain the fission products inside the cladding, (2) environmental resistance under normal operations (resistance to the hydrothermal corrosion and neutron irradiation) and (3) high-temperature oxidation resistance under a loss of coolant accident condition. This paper focuses on hydrothermal corrosion behavior of the SiC joints because of a significant lack of the knowledge. Stempien et al. reported in-pile corrosion behaviors of the SiC plate joints bonded utilizing TiC/SiC tape, calcium aluminate glass, or Ti foil [3]. All types of these joins were completely or partially debonded during the reactor coolant exposure for 44 days and 0.16 displacement per atom (dpa)-SiC. Therefore, development of the corrosion tolerant joint and the process method for SiC is a critical issue.

Various joining techniques utilizing different bonding phases are currently available for SiC materials. The candidate joining methods for the LWR cladding end-plug application can be converged based on the irradiation resistance. Katoh et al. demonstrated irradiation tolerant SiC joints fabricated by solid state diffusion bonding, Ti–Si–C reaction sintering based joining, SiC nanopowder sintering based joining, and glass-ceramic joining following neutron irradiation at 500 and 800 °C up to 5 dpa [4]. Those joining methods are potentially useful for the LWR pin end-plug applications in terms of the radiation resistance.

The solid state diffusion bonding typically utilizes an interlayer of a refractory metal such as titanium, molybdenum, and tungsten [5]. Metal silicides can be also used as the interlayer [6]. The joining method is well established and the advantage of this joining method is the relatively low joining temperature (~1500 °C), which

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can avoid the potential damage to the cladding, especially for SiC fibers in the composite form. The strengths of Hi-Nicalon Type S and Tyranno SA SiC fibers are known to degrade by annealing at ~1800 °C and ~1900 °C, respectively [7]. The drawback is the need for applied stress during the joining, which is typically more than a few MPa [5]. Jung et al. recently overcame this limitation by solid state diffusion bonding using a Ti interlayer and addition of Si powder with applied stress of only ~0.1 MPa [8]. The diffusion bonds were typically robust [4,9]. It may be possible to increase the environmental resistances of this type of the joint by choosing an appropriate interlayer.

The reaction sintering method utilizing the Ti–Si–C system yields a bonding layer of Ti_3SiC_2 (MAX phase) plus secondary phases including SiC, which was originally formed with applied stress of 30 MPa at 1200–1300 °C [10]. The pressureless joining method with Ti–Si–C system has been commercially available. The apparent shear strength of the joint was reported to be ~100 MPa [4]. The main concern of this type of the bonding layer is poor oxidation resistance of Ti_3SiC_2 phase [11], which may be improved by modification of the composition [12].

The SiC nanopowder sintered joint is formed by sintering mixture of SiC nano-powder and oxide additives such as alumina-yttria [13,14]. The additives form eutectic liquid during the heating, and then the liquid phase promotes sintering of SiC powder compact via the solution-precipitation process, which is commonly observed in liquid phase sintering [15]. If the eutectic liquid is a transient phase due to the evaporation, the joining method is specifically referred to as transient eutectic phase (TEP) sintering bonding [4,16]. Although the SiC nanopowder sintering typically requires relatively high processing temperature (~1850 °C) and applied pressure of ~10 MPa, this class of SiC joint exhibited the highest apparent shear strength among the different types of SiC joints [4]. The key development of this joint is a reduction of the joining pressure and control of the microstructure, especially for the secondary phases for improved environmental resistance [17,18]. The applied stress during sintering is potentially reduced by pressureless sintering [16].

The glass ceramics bond was demonstrated without applying an external stress at relatively low temperature (~1500 °C) for Ca–Al–O system [4,19]. The joint strength was typically ~100 MPa in torsion. Because of the discouraging results of the in-pile corrosion experiment on the glass ceramics joint [3], this joint was not investigated in this study.

The promising methods for the cladding end-plug application, which are not investigated in this study, are a selected area chemical vapor deposition (CVD) or infiltration (CVI) and a SiC forming polymer-derived joining [20], which can provide pure SiC bonding layer. Hybrid joining technique utilizing these two methods has been employed to the end-plug joining [21]. Hydrothermal corrosion behavior of high-purity SiC in simulated LWR coolant environments can be found elsewhere [22,23].

In this study, the corrosion behaviors of four types of SiC to SiC plate joints fabricated via the solid state diffusion with Ti and Mo interlayers, Ti–Si–C reaction sintering, and SiC nanopowder sintering were investigated under three types of LWR relevant chemical conditions, for up to five weeks for screening purpose. The corrosion experiment was conducted in autoclaves without radiation. Based on the corrosion rate and the microstructures of the corroded joints, possible methods to improve the corrosion resistance and future direction of the development of the SiC joints for the LWR cladding application were discussed.

2. Experimental

CVD β-SiC from Dow Chemical Co. (Marlborough, MA) with 99.9995% purity was used for the substrate material for all the joint specimens. The Mo diffusion bond was formed using a molybdenum foil (25 μm thick, 99.95% pure, Alfa-Aesar, Ward Hill, MA) as the bonding agent. The joining of SiC/metal/SiC sandwiches was accomplished by hot-pressing at 1500 °C, for 1 h, in flowing Ar-4% H_2 atmosphere, under a uniaxial pressure of 20 MPa. During the hot-pressing, presence of both hydrogen and titanium powder facilitated effective oxygen gettering. To form the Ti diffusion bond, a titanium foil (25 μm thick, 99.94% pure, Alfa-Aesar, Ward Hill, MA) was used as the interlayer. The joining of SiC/metal/SiC sandwiches was accomplished by hot-pressing at 1500 °C, for 1 h, in vacuum, under a uniaxial pressure of 17 MPa. For the Ti–Si–C reaction forming MAX phase bonding, a set of joining agent materials were purchased from Hyper-Therm High Temperature Composites, Inc. (currently Rolls-Royce High Temperature Composites, Inc., Huntington Beach, CA). The Ti–Si–C phase-based joints of CVD SiC were produced based on a pressure-less slurry process per the Hyper-Therm formula. Details of the raw materials and the process conditions are Hyper-Therm proprietary information. The SiC nanopowder sintered joint was fabricated using slurry of SiC powder mixture. To prepare the slurry, SiC nano-phase powder (average diameters ~30 nm, Nanomakers, France), and Al_2O_3 powder (average diameters ~0.3 μm, Kojundo Chemical Lab. Co., Ltd., Japan), Y_2O_3 powder (average diameters ~0.4 μm, Kojundo Chemical Lab. Co., Ltd., Japan) were dispersed in ethanol. The total amount of oxide additives was 6 wt% (weight ratio: Al_2O_3:Y_2O_3 = 3:2). The powders were mixed using milling with zirconia balls. The slurry was sandwiched by CVD SiC plates, and then dried at ~80 °C. After that, the SiC nanopowder sintered joint was formed by hot-pressing at 1850 °C, for 1 h, in an Ar atmosphere, under a pressure of 10 MPa. Note that the bonding layer contained the trace of zirconia from the milling media as shown in later. The processing conditions of the joints are summarized in Table 1. The bonded plates were machined into rectangular $12.0 \times 6.0 \times 1.5\ mm^3$ bars for the Mo and Ti diffusion and Ti–Si–C reaction sintered bonds and into square $6.0 \times 6.0 \times 3.0\ mm^3$ for the SiC nanopowder sintered joint. All the samples contained a hole with 2.0 mm diameter to hang on the specimens in autoclaves. The location of the bonding layers within the test specimens is shown in Fig. 1.

The hydrothermal corrosion experiments were conducted in 316 stainless steel autoclaves at GE Global Research Center (Schenectady, NY). The specimens were hung on a PTFE-insulated 304 stainless steel wire and immersed for 5 weeks in three LWR coolant environments, (1) pressurized water reactor (PWR) chemistry, (2) boiling water reactor (BWR) –normal water chemistry (NWC), and (3) BWR–hydrogen water chemistry (HWC) conditions. The details of test water chemistry conditions are summarized in Table 2. The PWR and BWR–HWC conditions were simulated reducing environments, while the BWR-NWC condition was representative of in-core oxidizing water chemistry in BWR. The water chemistries were controlled by injection of appropriate mixtures of argon, oxygen, and hydrogen. The water flow rate during the tests was 200 ml/min. One specimen of each type of the joint was exposed to each water chemistry condition. The specimen of the SiC nanopowder sintered joint for the PWR condition was absent. The samples were removed from the test autoclaves after two weeks for examining the stability of the joining areas, and then put them back to the test autoclaves again for an additional three weeks. Details of the experimental setup were reported elsewhere [22]. Additives used in the coolant of commercial LWRs such as Li, B, and noble metals [24] were absent in water of the autoclave. Such additives may affect the chemical/physical stability of the joining area [24].

The recession of the SiC substrate was evaluated from weight loss of the rectangular specimens, ignoring the bonding layer, because volume of the bonding layer within the corrosion