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Microstructural and thermal property evolution of reaction bonded silicon carbide (RBSC)

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ABSTRACT

Temperature dependent thermal conductivity of reaction bonded silicon carbide (RBSC) from 300 K to 1073 K is evaluated. The thermal conductivity of 80 vol% and 90 vol% SiC RBSC is measured to be 185.7 W/m·K and 211.4 W/m·K at room temperature and decreases to 51.46 W/m·K and 55.77 W/m·K at 1073 K, respectively. Thermal transport behavior of RBSC at elevated temperatures suggests that a structural mechanism plausibly associated with phase transformation occurs in the composite system. TEM *in-situ* heating test is employed to investigate the RBSC phase evolution. Results indicate the existence of minor Si amorphous phase near SiC/Si interface, and the amorphous Si phase transformation to crystalline Si could start at a relatively low temperature. The Si phase transformation improves the thermal transport performance of the RBSC system.

1. Introduction

Ceramic composites have replaced alloys in various thermal and energy related applications due to their remarkable thermal properties and high environmental stability at extreme conditions [1]. Silicon carbide based composites are premier candidates in many high-tech fields, not only for advanced properties as mentioned above, but also for their outstanding structural properties such as light weight and high stiffness [2]. For heat exchanger [3] and electro-static chuck [4] applications, the thermal conductivity of ceramic composites is a crucial property and continues to be an active research area. Indeed, the thermal conductivity of SiC based composites has been studied extensively [5–8], but few of them can reach 200 W/m·K and is also significantly below that of the single crystal SiC [9] due to the multi-phase nature of the composites. The reduction of thermal conductivity in SiC composites is primarily attributable to the thermal resistance introduced by grain boundaries, secondary phases, impurities or additives, and other structural defects.

There has been great interest in understanding and improving the correlation of microstructure and thermal conductivity of ceramics and ceramic composites. Aghajanian et al. [10] studied the effect of grain size on thermal conductivity of reaction bonded silicon carbide (RBSC) composites, and showed that when the average SiC grain size increased from 6 to 50 μm, the thermal conductivity increased from 137 W/m·K to 188 W/m·K at room temperature. Sigl [7] indicated that the existence of $Y_3Al_5O_{12}$ oxide phase (YAG) could significantly decrease the thermal conductivity of liquid phase sintered SiC composites due to the inclusion of YAG, a phase of extremely low thermal conductivity. Yu et al. [11] further indicated that the distribution of Y_2O_3 secondary phase in AlN matrix reduced composite thermal conductivity because the minor phase would disrupt AlN grain connection by forming an amorphous layer.

To date, there has been limited research focusing on the temperature dependence of the RBSC thermal properties even though RBSC composites have been widely utilized in different industrial fields with strict requirements on the thermal properties [12–14]. Understanding the correlation of microstructural and thermal properties of RBSC and their evolution is therefore essential to expanding their applications in advanced technologies. Moreover, the same knowledge is anticipated to contribute to optimizing the

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fabrication parameters of RBSC and developing novel microstructures and applications.

2. Materials and methods

For thermal property measurement, cuboid samples of 10 mm × 10 mm × 5 mm were cut from 80 vol% SiC and 90 vol% SiC RBSC composites (M cubed Technologies, Newark, Delaware, USA). Density (ρ) was calculated by dividing sample weight by the volume obtained using the method of water immersion in a graduated cylinder. The front and rear faces of each sample were polished on P240 SiC papers (Buehler), followed by Pt and carbon coating. The thermal diffusivity (α) and heat capacity (C_p) were measured in the temperature range from 300 K to 1173 K by the laser-flash technique [15] using a DLF-1200 instrument (TA Instruments, Newark, Delaware, USA) with the thermographite as a reference material. Thermal conductivity (k) was calculated based on measured α, C_p and ρ according to $k = \alpha \times C_p \times \rho$.

The phase composition of 80 vol% and 90 vol% SiC RBSC was determined by using a Bruker D8 X-ray diffraction (XRD) instrument. SEM was performed with a Zeiss Auriga 60 focused ion beam and scanning electron microscope (FIB/SEM) equipped with Oxford X-Max 80 X-ray energy dispersive spectroscopy (EDS). TEM *in-situ* heating specimen was prepared by a FIB lift-out approach. The electron transparent specimen containing the SiC/Si interface was attached to a heating chip (E-AHBN model, Aduro E-chips, Protochips™) by localized Pt deposition (welding). *In-situ* heating test was performed in a JEM-2010F transmission electron microscope (TEM) operating at 200 kV with an Aduro 300 heating sample holder. The temperature was adjusted from room temperature to 1173 K at a nominal rate of 1 K/s.

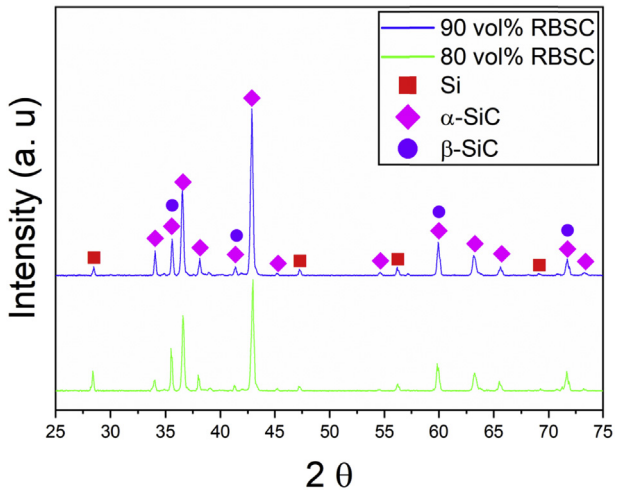

Fig. 1. XRD patterns of the 80 vol% and 90 vol% SiC of RBSC composites.

3. Results

3.1. Microstructure

The XRD patterns in Fig. 1 reveal that the composite contains α-SiC, β-SiC and Si. The α-SiC phase includes the original SiC grits in the preform and those formed during the reaction bonding fabrication. Based on a "carbon dissolution and saturation" mechanism [16,17], the carbon atoms in molten Si moves to cold spots (pre-existing SiC) due to carbon concentration and temperature gradients during the infiltration, and it then becomes saturated to form "new" SiC with cubic structure (β-SiC). Those newly formed SiC near reaction front can further transform to α-SiC due to an elevated local temperature in the reaction zone.

The SEM of RBSC appears to include two major phases, SiC and Si, as is consistent with the XRD results. Fig. 2a shows the typical microstructure of RBSC containing pre-existing SiC (dark-grey), newly formed SiC (light-grey), and residual Si (grey). It is suggested that the contrast is due to the different impurity levels, in which the pre-existing SiC contains less impurities (such as Al), thus appear dark [16]. The distribution of SiC and Si is mapped based on C and Si characteristic X-ray signals, as shown in Fig. 1b. A selected area electron diffraction (SAED) pattern (upper-right) shown in Fig. 3a indicates that the SiC has a cubic structure (β-SiC) and overall, crystalline Si is distributed as trunks and a network in between other grains. However, at sporadic locations and interfacial regions, small volume of amorphous Si (a-Si) exists as suggested by the SAED in the lower-right inset of Fig. 3b. The small volume of a-Si can be introduced by a thermal stress that, together with an abrupt local temperature drop as the molten Si is in contact with bulk α-SiC, suppresses the periodicity of the Si. In the complex composite system, when there is a temperature gradient, phases generate varied stress due to thermal expansion coefficient difference (i.e., at room temperature, $2.7 \times 10^{-6}K^{-1}$ for 3C-SiC [18]; $3.85 \times 10^{-6}K^{-1}$ for 6H-SiC along c-axis [19]; $2.57 \times 10^{-6}K^{-1}$ for Si [18]). The existence of atomically non-periodic structure can significantly affect the thermal transport properties of RBSC. In this study, the structural characterization is focused on investigating the interfacial details between SiC and Si.

3.2. Thermal properties

The density of 80 vol% and 90 vol% SiC RBSC was measured and calculated to be 2.965 g/cm^3 and 3.066 g/cm^3 respectively at room temperature. The temperature dependence of their thermal

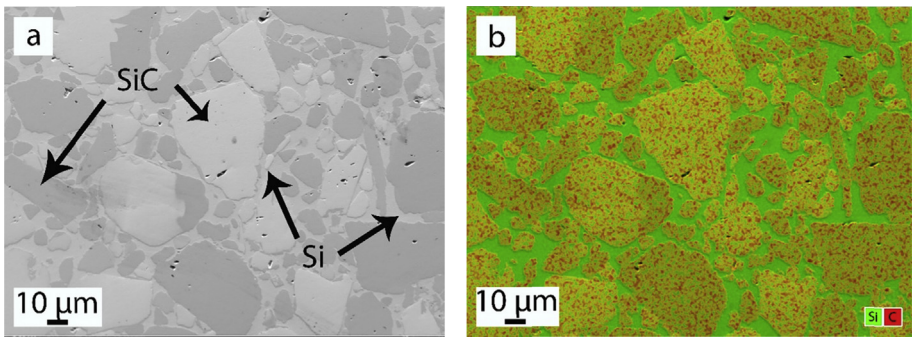

Fig. 2. SEM image and EDS map of RBSC composite: (a) SEM image captured by secondary electron detector, (b) Overlay of EDS maps of a polished RBSC surface.