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Short communication

Thermal conductivity studies on Si/SiC ceramic composites

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Abstract

Ceramic heat exchangers are increasingly used in many nuclear power plants. Silicon carbide has been treated as a promising material for heat exchanger application since it has good thermal conductivity and corrosion resistance. In this work, four different types of Si/SiC ceramic composites were prepared by liquid silicon infiltration technique. Thermal conductivities of these ceramic composites at different temperatures are measured by the laser flash thermal conductivity method. Results show that the presence of free carbon and voids are notably affecting the thermal conductivity of these materials.

Keywords: B. Porosity; C. Thermal conductivity; D. SiC; Ceramics

1. Introduction

Silicon carbide ceramics possess outstanding mechanical, thermal and electrical properties. Hence these are widely used in structural, nuclear and semiconductor applications. Its high stability with respect to neutron radiation, associated with sufficient thermal conductivity, gives scope for a potential application in nuclear industries [1,2]. Recently, SiC has become a prime candidate material for high-temperature heat exchanger applications, primarily because of its excellent thermal stability and corrosion resistance in severe environments, and sufficient thermal conductivity at elevated temperature [3]. In general, plate type and tube type heat exchangers are used in various power plants. Various manufacturing techniques have been used to produce SiC for heat exchanger application. Hofenauer et al. [4] used wood powder as a raw material for producing Si/SiC ceramic, for a plate type heat exchanger. The main advantages associated with this process are low cost, renewable raw material (i.e. wood), easy fabrication of complex shapes, near net-shape formation and isotropic nature of the final product. Similarly other researchers produced Si/SiC ceramics from cotton fabric, jute fiber, etc. [1–3]. When the material is employed as a heat exchanger component, it is important to study the specific heat, thermal diffusivity and thermal conductivity of the materials of interest before being used. Even though these Si/SiC ceramic composites are considered as a potential material for heat exchanger application, the thermal conductivity behavior of these materials has not been well studied. Hence, the aim of the present work is to determine the thermal conductivity of the Si/SiC ceramic composites and to study the influence of microstructure on the thermal behavior. The laser flash thermal conductivity method has been used to assess the thermal conductivity of the above ceramic composites. The microstructures of the ceramic composites have been analyzed using scanning electron microscope.

1.1. Theory of thermal conductivity

Thermal conductivity, K, is the intrinsic property of a material that indicates its ability to conduct heat. In contrast to metals, in which electrons carry heat, most ceramics transport heat primarily by phonons. Phonon is a quantum of vibration. Every harmonic vibration can be decomposed into elementary vibrations called phonons [5]. The total number of phonons in a system that vibrates (e.g. crystal) is related to the temperature of the system. At higher temperatures, vibration of an object is stronger and the number of phonons larger.

Phonon–phonon interaction plays an important role for the thermal conduction in most of the ceramics. In a perfect crystal, the transport of phonons will be scattered easily by other phonons on increasing the temperature. However, in real dielectric solids,

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Fig. 1. Thermal conductivity samples.

imperfections such as pore, impurity, crystal boundary and dislocation, interrupt with phonons causing phonon scattering. Hence the mean free path of phonon drastically comes down and it can be described by Matthiessen Rule [5,6]:

$$\frac{1}{\lambda} = \frac{1}{\lambda_{defect}} + \frac{1}{\lambda_{boundary}} + \frac{1}{\lambda_{phonon}} \quad (1)$$

So, the thermal conductivity of the dielectric materials is given by

$$K = \frac{\rho \lambda \upsilon C_p}{3} \quad (2)$$

where ρ is the bulk density of the solid (in g/cm^3), C_p is the heat capacity of the solid (in J/g K) and υ the elastic wave velocity in the solid (in cm/s). Instead, one can also easily determine the thermal conductivity of the materials by using the following equation:

$$K = \rho C_p \alpha \quad (3)$$

where α is thermal diffusivity of the solid.

2. Experimental procedure

Wood powder based Si/SiC ceramic composites were produced by mixing two different sizes of wood powders (−200 μm and +1000 to −1200 μm) with 40% phenolic resin individually. For homogeneous mixing of binder with wood powder, the phenolic resin was diluted with isopropyl alcohol. Subsequently, phenolic wood mixtures were hot pressed in a metal mold to a dimension of 100 mm × 100 mm × 9 mm at 160 °C. Carbon preforms were prepared by pyrolyzing the dried (100 °C, 10 h) wood powder composites at N_2 atmosphere in a tubular furnace. A heating rate of 2 °C/min was used up to 600 °C and a heating rate of 5 °C/min was used up to 1100 °C. The resulting porous carbon preform was infiltrated with liquid silicon in vacuum graphite furnace at 1600 °C for 2 h. Preparation of cotton fabric based plate type component is discussed elsewhere [1,6]; where the plate components were produced by hand lay-up process using phenolic resin as a binder. Samples produced are denoted as FW (fine wood particle Si/SiC ceramics), CW (coarse wood particle Si/SiC ceramics), CT (cotton fabric based Si/SiC ceramics) and CTCVI (cotton fabric based chemical vapor infiltrated (CVI) Si/SiC ceramics). The microstructures of the above ceramic composites were observed with a scanning electron microscope (FEI, QUANTA 200)

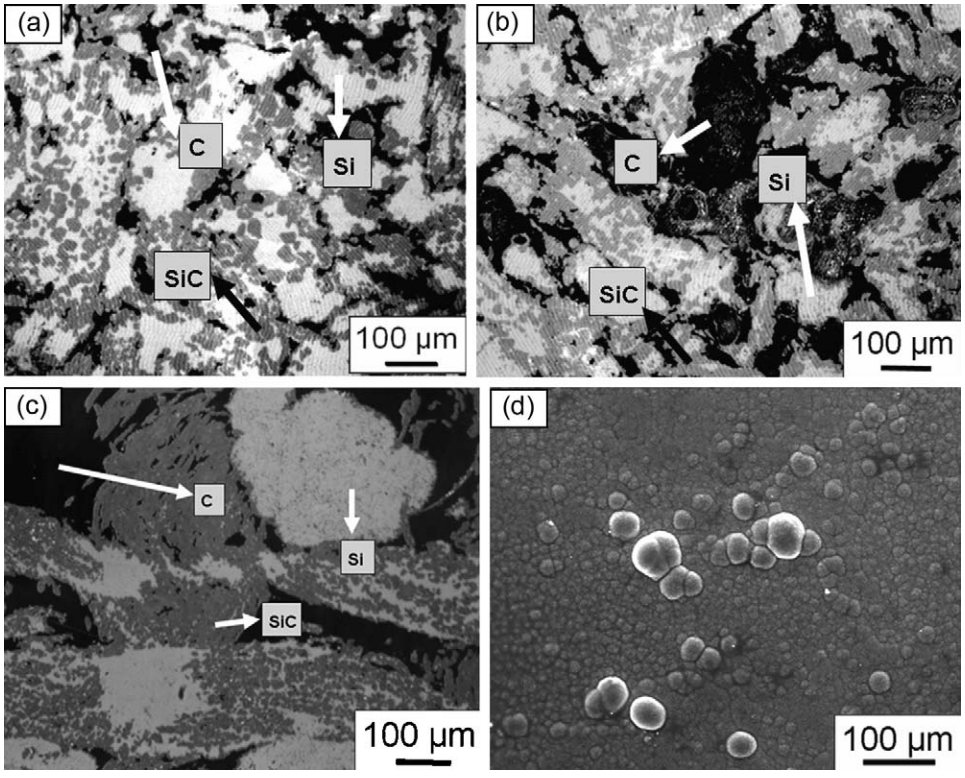

Fig. 2. SEM micrographs of the surface of (a) FW, (b) CW, (c) CT and (d) CTCVI ceramic composites.

operated at 30 kV and 20 mA. Density was determined by the Archimedes method.

For measuring thermal properties, disks of 10 mm in diameter and 4 mm thick were cut from the Si/SiC composite plates. Both the sides of the disc were mirror polished with the help of 1 μm diamond particle. Before measuring the thermal conductivity, the surface was coated with a layer of gold to 60 nm thickness followed by a coating of carbon black. Thermal conductivity measurement was carried out by laser-flash method (Flash sline 5000 Thermal diffusivity system-Anter Corporation, U.S.A). The thermal conductivity (K) was calculated by using the Eq. (3). The thermal conductivity samples are shown in Fig. 1.

3. Results and discussion

Fig. 2 shows the surface morphology of Si/SiC ceramic composites. The light gray areas are silicon carbide and the dark regions are free carbon. Free silicon is also present in some of the retained pores and it appears white. These phases are confirmed by the EDAX analysis. The properties of the ceramic composites are given in Table 1.

Table 1
Properties of Si/SiC ceramic composites.

Samples	Density (g/cm^3)	Porosity (%)	SiC[a] (%)	Si[a] (%)	C[a] (%)
CT	2.5	23	60	22	18
CTCVI	2.8	14	–	–	–
FW	2.9	11	76	14	10
CW	2.7	19	69	17	14

[a] Based on EPMA surface mapping.

Fig. 3 shows the variation of diffusivity, specific heat and thermal conductivity as a function of temperature. The thermal conductivity was calculated by multiplying the thermal diffusivity, specific heat and room-temperature bulk density. The thermal diffusivity values decrease over the entire temperature range. The specific heat increases, as can be expected from the Debye theory [7–9]. The inverse temperature dependence of diffusivity of the above SiC ceramics is suggesting a dominant phonon conduction behavior, which resembles most polycrystalline materials [8]. The thermal conductivity also decreases over the entire temperature range (Fig. 3b). This value is much lower than the reported room temperature thermal conductivity of liquid phase (110 W/mK) and hot pressed SiC (120 W/mK) ceramics [10]. However, this

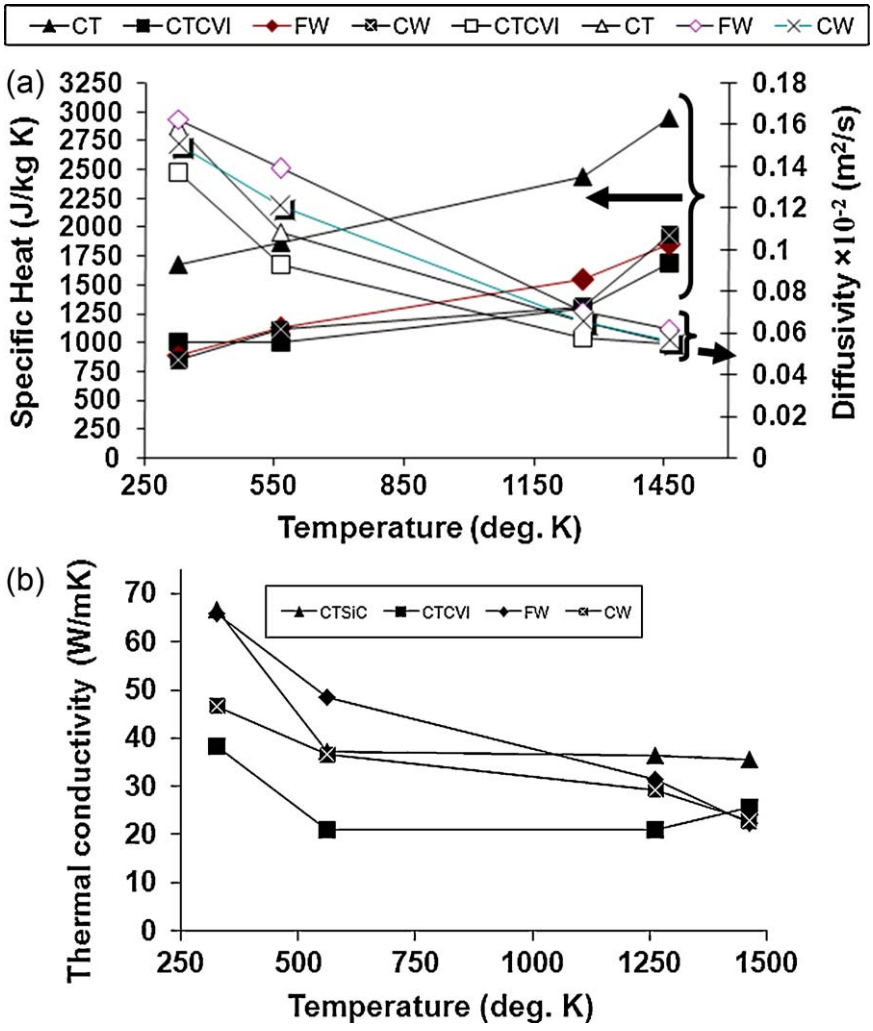

Fig. 3. Thermal properties of Si/SiC ceramic composites: (a) specific heat and diffusivity and (b) thermal conductivity.

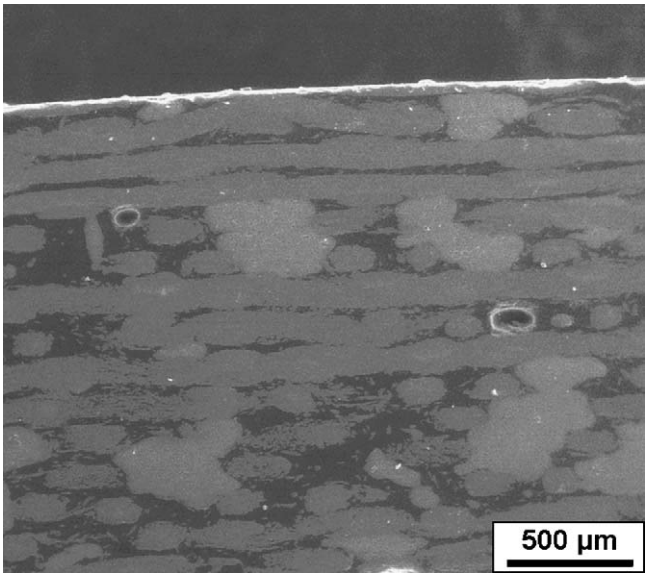

Fig. 4. SEM micrograph at the cross section of the CT composite.

value is much better than the thermal conductivity exhibited by the commercially available CVI-SiC/SiC composite [11]. There are three main reasons for the low thermal conductivity behavior of the above Si/SiC composites. First reason is the presence of glassy carbon. The SiC fibers/grains are covered by glassy carbon and it is a poor thermal conductor. The thermal conductivity of glassy carbons in the range 0.1–300 K is probably dominated by the presence of voids [8]. Even though the thermal conductivity is similar to that of a glass, any localized excitations present must have a minor influence on thermal transport. In general, glassy carbon may have two different size pores, i.e. submicroscopic voids and large voids (10^{-4} cm). Depends on the void size and temperature, the phonon mean free path may vary from Å to 10^{-3} cm. Hence, there may be a chance of high phonon scattering within the existing free carbon and that leads to low thermal conductivity [8,11–13]. Second, the direction of the fiber with respect to the heat flux (especially in CT and CTCVI samples). The absence of fibers in the third direction (i.e. parallel to the heat flux) results in anisotropic nature. For thermal conductivity measurement, samples were prepared from the plate type composites, where all the fibers are perpendicular to the heat flow direction (Fig. 4). Third, the phonon velocity varies with respect to the nature of materials. Therefore, there may be large chance of scattering at the interface of the two different materials [14,15]. Since the Si/SiC composite materials are composed of three different phases, such as SiC, Si, and glassy carbon, they are hindering the phonon movement. This can lead to low thermal conductivity. Apart from these reasons, the pores present in these composites can also act as a barrier for thermal conductivity. The thermal conductivity of the CVI treated SiC composite (CTCVI) is significantly lower than the untreated SiC (CT) composites. Commonly, fine SiC crystallites are produced during chemical vapor deposition. When the crystallite sizes are fine, the mean phonon free path drastically comes down due to the scattering at the boundaries. This significantly affects the thermal conductivity of the materials [8,9]. Fig. 2b also shows a slight increase in the thermal conductivity of CVI treated samples beyond 1000 °C. This may be due to the coarsening of the SiC crystallites at that high temperature [10,14]. Even though the properties of the CW samples are better than the CT samples (Table 1), lower thermal conductivity may be due to the finer SiC grains surrounded by the glassy carbon.

4. Conclusion

Low thermal conductivity of the Si/SiC ceramic composites may arise from the presence of voids and excess glassy carbon surrounding the SiC grains. Therefore, further developments, such as the removal of excess free carbon, avoiding void formation during the manufacturing process and the incorporation of fibers in the thickness direction in the Si/SiC composites are necessary to increase the thermal conductivity.

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