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Effect of Gradient TiC–Ti_5Si_3–$TiSi_2$ barrier layer on SiC in SiC–borosilicate glass composites

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ABSTRACT

The oxidation of SiC in SiC–borosilicate glass composites releases gases which cause a high volume expansion ratio and low bending strength and limits the application of the composites as packaging materials. To solve this problem, we deposited a Ti compound coating on SiC by heating a mixture of TiH_2 and SiC particles at 750 °C for 1 h under vacuum. The thickness of the coating was about 200 nm, and the coating was a gradient TiC–Ti_5Si_3–$TiSi_2$ barrier layer according to the result of the X-ray diffraction. The coated SiC had a better anti-oxidation ability than uncoated ones in borosilicate glass, and as a result, the volume expansion ratio of the composites with 25 vol.% coated SiC was reduced by 287%, and the bending strength was increased by 117%. The composites with 55 vol.% SiC had the maximum thermal conductivity (14.2 $Wm^{-1}K^{-1}$) and rather low thermal expansion coefficient (4.98×10^{-6}).

1. Introduction

With the development of electronic technologies, electronic equipments have greater promotion in power density meaning that packaging materials with high thermal conductivity are necessary for electronic equipments. Metal based packaging materials such as diamond/Al, diamond/Cu, SiC/Al, and SiC/Cu have been extensively researched [1–4]. However, metal-based packaging materials have much higher coefficient of thermal expansion (CTE) than electronic equipment (Si). Recently, glass based packaging material has been observed due to its low density, low sintering temperature and close CTE with electronic equipment [5,6]. As SiC has better wettability with glass than diamond, SiC is preferentially used in glass based packaging materials. However, SiC particles in the SiC-glass composites are seriously oxidized at elevated temperatures and the reactions release gaseous resultants such as SiO, CO, and CO_2, which form pores in the composites and cause extremely low bending strength and thermal conductivity [7,8]. Therefore, the electron industry must find a method to avoid the oxidation of SiC in glass.

It is known that SiC has excellent anti-oxidation properties in air due to the protecting SiO_2 layer on the surface of SiC. According to the researches of Yang [9], Vaughn [10] and Shi [11], SiC only becomes distinctly oxidized when it is heated at a temperature higher than 1600 °C or heated in a low oxygen pressure environment. However, in the SiC–borosilicate glass SiC was seriously oxidized at temperatures lower than 850 °C. According to the research of Jackson [12] and Dey [13] and our previous research [14], the SiC in SiC–borosilicate glass composites was oxidized due to the effect of the oxygen and the alkali oxides (R_2O). If oxygen or R_2O lacked, the oxidation would stop. The oxidation reactions are listed in Eqs. (1), (2) and (3).

$$SiC + O_2 \rightarrow SiO_2 + CO_2 \quad (1)$$

$$SiC + O_2 \rightarrow SiO + CO \quad (2)$$

$$SiO_2 + R_2O \rightarrow R_2SiO_3 \quad (3)$$

Our previous research [14,15] added Ti or Zn elements to absorb the oxygen in composites to protect SiC, but the large amount of oxidation resultant of Ti or Zn changed the composition of borosilicate glass seriously, which dramatically raised the refractoriness of composites. Depositing a barrier layer on surfaces of SiC particles would restrict the oxidation of SiC in glass by isolating SiC from O_2 and R_2O. Villegas [16] thickened the SiO_2 layer through oxidation treatment, but this treatment caused a degradation of SiC and is not useful for fine particles. Magnani [17] and Ramasany [18,19] used a rare earth oxide coating on surface of SiC ceramic composites to resist oxygen from the air, but this method could not apply uniform coatings on SiC particles and some SiC particles were still oxidized.

In this paper, we deposited a gradient TiC–Ti_5Si_3–$TiSi_2$ coating on SiC particles using a thermal reaction method. This coating was expected to

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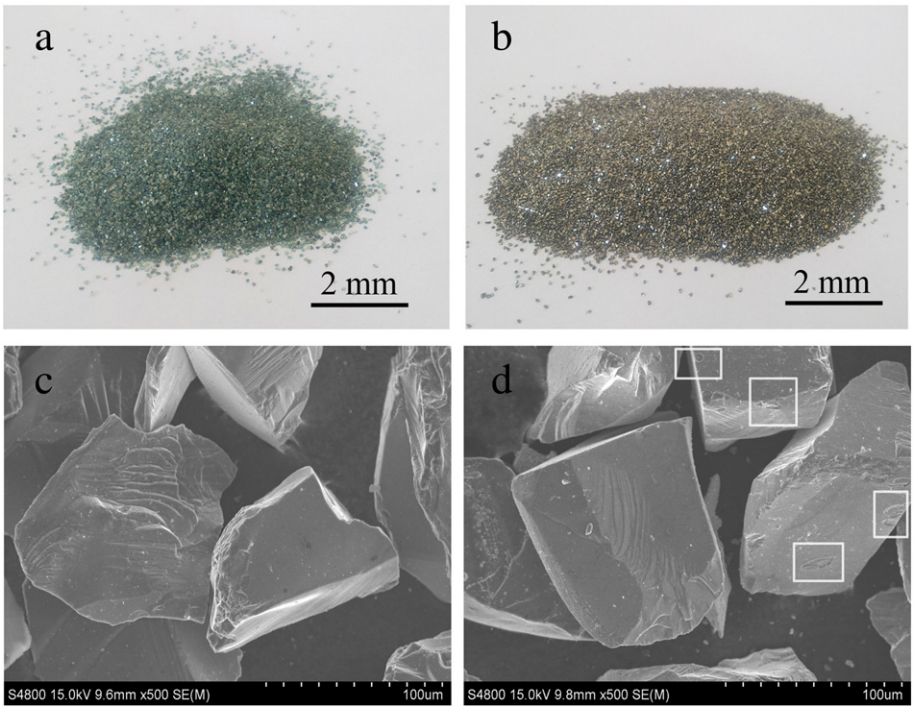

Fig. 1. Digital photos of uncoated SiC (a) and TiC–Ti_5Si_3–$TiSi_2$-coated SiC (b) particles; FESEM images of uncoated SiC (c) and coated SiC (d).

isolate the SiC from oxygen and alkali oxides. In addition, the oxidation resultants of the coating were TiO_2 and SiO_2, which were network-forming oxides in glass, so it was expected that the coating had good wettability with glass [20–23]. The properties of the coating were studied and the performances of the coating in SiC-glass composites were researched. The thermal conductivity and CTE of the composites with different contents of SiC were investigated.

2. Experimental

In this research, α-SiC particles (with a mean size of 75–90 μm) were used. TiC–Ti_5Si_3–$TiSi_2$-coated SiC was prepared through a thermal reaction method. Firstly, SiC particles were uniformly mixed with TiH_2 powders (with a mean size of 1–5 μm). The mixture was kept in a vacuum reactor under 10^{-3} Pa for 1 h, and then heated at 750 °C for 60 min.

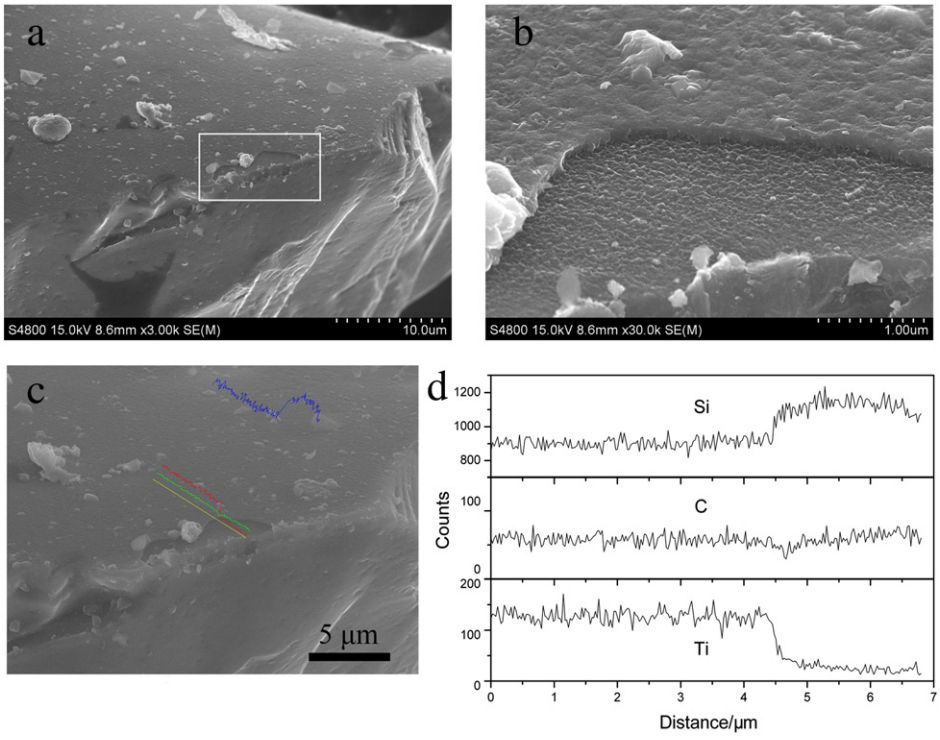

Fig. 2. Morphology and element distribution of coating, a coated SiC with a small area of surface peeled off (a), magnifying image of the circled surface (b), EDS line scan on coated SiC (c), the EDS results (d).