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Research on Reaction-bonded SiC for Optical Application

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Abstract: Ultrasonic treatment technology is utilized to fabricate RBSiC, and a new polishing technology– float polishing is developed to process RBSiC. SEM, XRD, AFM. Raman scattering and WYCO are applied to measure and analyze the RBSiC and the polished surface. The results indicate that the ultrasonic treatment decreases the content of the free Si and makes RBSiC denser and more uniformity. The float polishing technology is suitable for polishing RBSiC effectively.

Key words: ultrasonic treatment; green body; reaction bonding; float polishing; infiltrating
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1 Introduction

Silicon carbide (SiC) is a good ceramic material for optical application because of its many attractive properties such as low density, low thermal expansion coefficient, high thermal conductivity and high thermal resistance, etc. SiC was used as the replacement Be and glass in water-cooled laser mirrors during the decade of the 1980's, and it has been used to fabricate lightweight space mirrors since 1986.

SiC is a family of materials in several forms, which are conveniently classified by processing method. The primary processing methods involve (1) hot pressing SiC powders, (2) formation by deposition, (3) reaction bonding of a silicon carbide green body with molten silicon.

1. Hot pressed SiC

Hot pressing produces the highest strength and highest modulus SiC. Micrometer-sized SiC powders are combined with suitable additives, which serve as hot pressing aids and grain growth inhibitors. The resultant product is near 100% dense α-SiC which exhibits exceptional mechanical properties. The combination of high pressure and temperature required limits the size range of optical structures and fabrication feasibility. The current commercial material is extremely difficult to be processed. However, impurity additives result in decreased thermal conductivity and other chemical properties. The major limitation for hot pressed SiC is the inability to form near net shape complex configurations, which are required to lightweight mirrors and optical structures.

2. Chemical vapor deposited (CVD) SiC

Chemical vapor deposited (CVD) SiC is a 100% dense β-SiC. The process uses a vacuum chamber between 1275– 1350℃. The silicon carbide particles are deposited on a graphite substrate. The basic microstructure is polycrystalline modular and can be polished to < 1.0nm RMS. The material is extremely hard and requires both diamond tools and polishing compounds to polish. The advantage of CVD silicon carbide is its high radiation survivability when it's not deposited silicon rich. The capability to deposit 0.50-inch thick substrates has been demonstrated. However, this process is limited in the size of substrates and is also very expensive.

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3. Reaction-bonded SiC(RBSiC)

The process approach involves generation of a green body of silicon carbide. Generially, the green bodies are fabricated by slip casting, and are presintered prior to infiltration with silicon. The infiltration process is carried out at a temperature significantly higher than the silicon melting point since bonding of the silicon carbide grain is the key to developing good mechanical properties. These procedures produce dense microstructure consisting of a bonded network of silicon carbide with isolated regions of free silicon comprising ~ 30% . The two-phase microstructure also presents some advantages in other fabrication areas.

All the three process approaches produce different microstructures and have been used successfully to make optics, in which, the hot pressed SiC has excellent Weibull strength but is difficult to form into complex shapes, the CVD SiC is the most polishable but is limited by deposition thickness, and the RBSiC is amenable to complex shapes but difficult to polish cost-effectively. The applications has demonstrated that RBSiC has the most flexibility.

The properties of RBSiC can be tailored by controlling the grain size and the content of free Si, and the uniformity of the microstructure also influences the properties of RBSiC. In this work, we provide a new method to densify the microstructure and decrease the content of Si in order to obtain RBSiC with better properties[1-6].

2 Experiment

The first step is to fabricate green body : making a slurry of α-SiC grains, carbon-rich resin and additives. During molding and casting, the ultrasonic treatment technology is utilized. SiC grain size is about 1μm~ 10μm.

The second step is Si infiltration: carbonizing the green body, then firing it at the Si atmosphere (at 1650℃) . By this step we obtain RBSiC.

The third step is to process RBSiC. The RBSiC sample is ground and polished. The surface is first ground by SiC for 8h and polished by Al_2O_3 for 8h, and then float polished for different periods (Fig. 1).

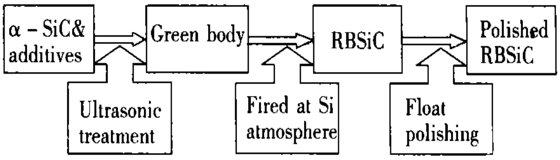

Fig. 1 Flow chart

All the samples are measured and characterized by SEM, XRD, AFM and WYCO.

3 Result and discussion

3.1 Characterization of RBSiC

Fig. 2 is the SEM photo of the fracture section, which indicates that the RBSiC body is dense; there aren't any pores and microcracks. Measured by a suspending method, we obtained the density of the RBSiC. The density is about 3.09g/cm3, which is higher than the reported results. Fig. 3 is the XRD scheme of the RBSiC. These peaks are corresponding to the XRD positions of α-SiC, β- SiC and Si. Calculating from the scheme , the content is 9% for Si, 11% for β- SiC and 80% for α-SiC, approximately.

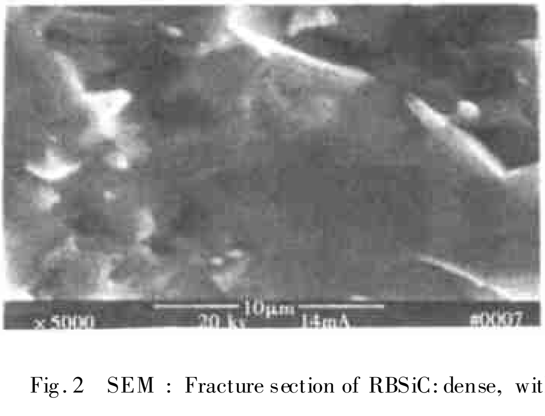

Fig.2 SEM : Fracture section of RBSiC: dense, without pores and cracks

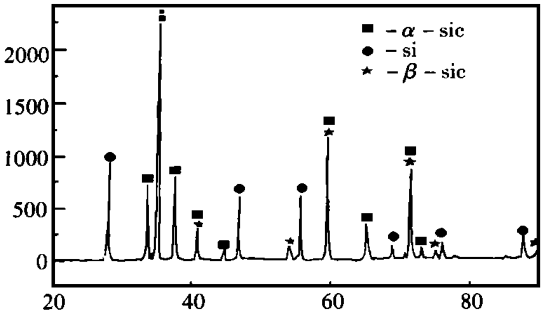

Fig. 3 X-ray diffraction of RBSiC

3.2 Effects of ultrasonic treatment

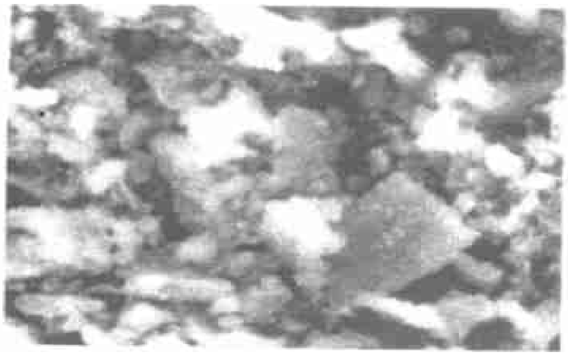

Fig. 4 SEM: Fracture section without the ultrasonic treatment(× 2000)

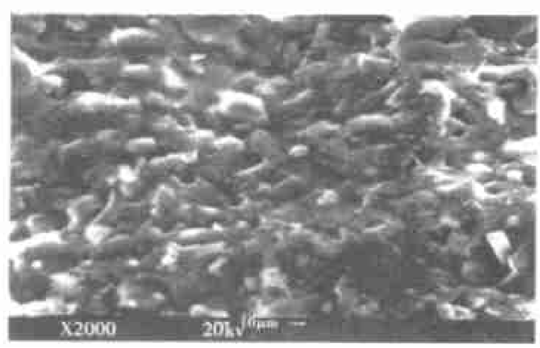

Fig. 5 SEM: RBSiC without the ultrasonic treatment (× 2000)

The ultrasonic treatment plays a great role in the fabricating process of RBSiC. Fig. 4 shows the microstructure of the green body formed without ultrasonic treatment. There are large pores in the body, the grains contact each other in a loose way, which results in the formation of pores when such a green body is reactively bonded (Fig. 5). When the ultrasonic treatment technology is applied, the green body is formed in a dense way. The grains are combined closely, and the gaps between the grains become smaller than that in the green body formed without the ultrasonic treatment (Fig. 6-7). After reactively bonded, such a green body is infiltrated thoroughly by Si (Fig. 2).

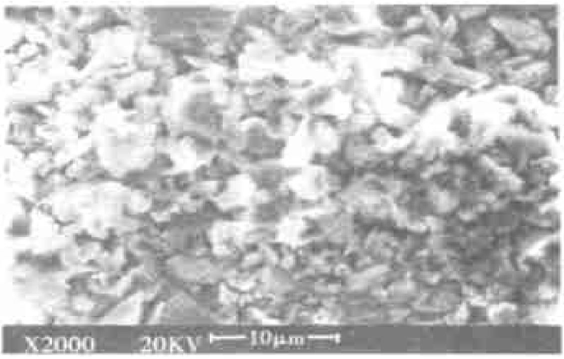

Fig. 6 SEM: After the ultrasonic treatment (× 2000)

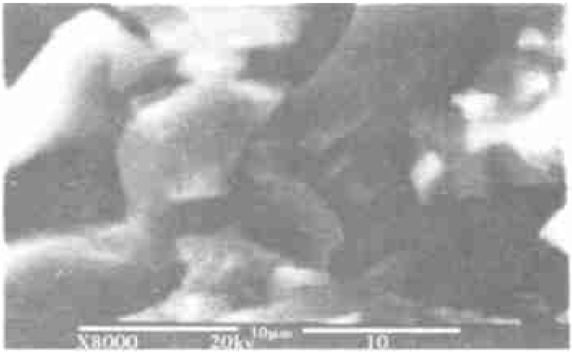

Fig. 7 Zooming in: SiC grains covered and combined by carbon(× 8000)

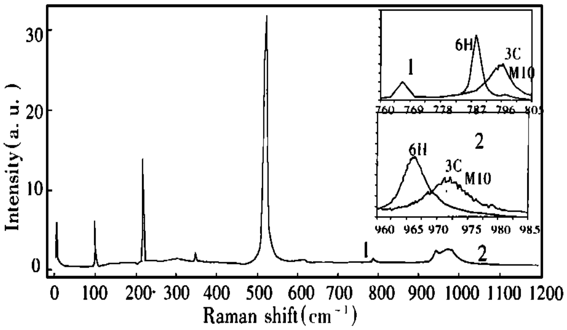

Fig. 8 Micro-Raman scattering of RBSiC: local resolution of peak 1 and 2

3.3 Micro-Raman scattering

In Fig. 8, Raman spectra of α-SiC, β-SiC and Si are shown. In case of β-SiC, two peaks can be observed, one is at 972cm^{-1} which can be assigned to longitudinal optic(LO) phonon peak, the other is at $796cm^{-1}$ which is assigned to transverse optic (TO) phonon peak. Also there are some weak peaks in addition to LO and TO phonon peaks. These peaks are supposed to be caused by some disorder or defects in the crystal.

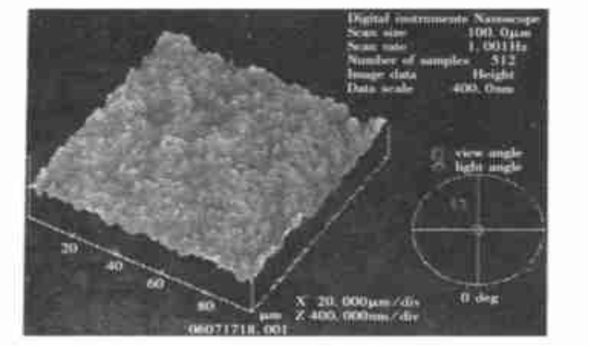

Fig. 9　AFM : ground for 8h(using SiC) then polished for 8h(using Al_2O_3)

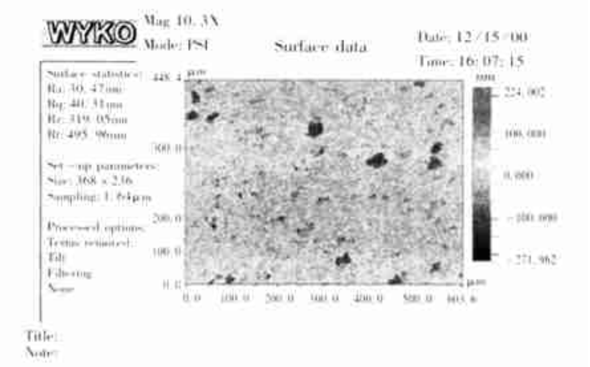

Fig. 10　WYCO: polished for 8h(using Al_2O_3)

α-SiC (including 4H and 6H) shows strong peaks at the region of $100cm^{-1}$ ~ $300cm^{-1}$(TA region), $760cm^{-1}$ ~ $800cm^{-1}$ (TO region) and $965cm^{-1}$ ~ $975cm^{-1}$ (LO region). Expanded spectra of TO and LO region are shown in the up-right. At the TO region, 6H shows a weak peak at the same position as β-SiC($796cm^{-1}$) and the peak of 6H is located at $789cm^{-1}$. At the LO region, we can also distinguish spectra of β-SiC from α-SiC(6H)[7-8].

3.4　Polishability of RBSiC

Fig. 9 is the AFM of the surface ground by SiC and polished by Al_2O_3 for 8h, respectively. Because of the hardness difference between Si and SiC, the Si part in the RBSiC body is removed easier than SiC. All the "mountains" left on the surface are SiC grains, so we can conclude that the RBSiC body consists mainly of SiC. Fig. 10 is WYCO result of the said surface, Ra is about 35nm, which shows RBSiC is difficult to be polished by traditional methods. Fig. 11-Fig. 12 show the said surface polished by another unique way–float polishing. In the course of the float polishing, the surface quasi contacts with the polishing pan. With the spin of the pan, polishing liquid polishes the surface uniformly. After 12h float polishing, Ra is about 3.4nm, which shows that the float polishing is suitable to process the bimodal medium such as RBSiC.

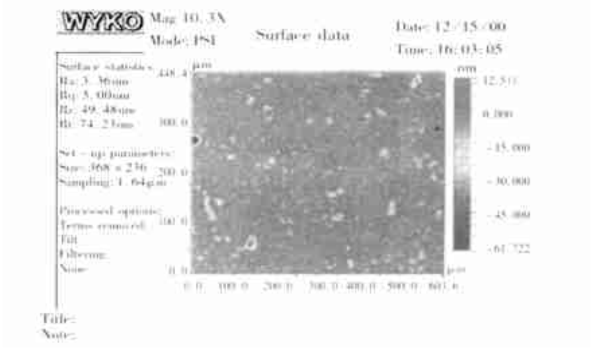

Fig. 11　WYCO : float polished for 12h

Fig. 12　Surface after the float polishing (30mm)

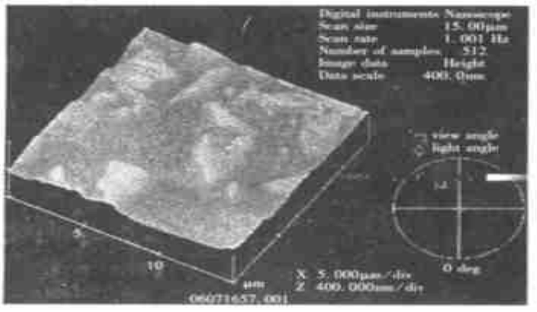

Fig. 13　AFM: Existing form of β-SiC and its combination with α-SiC

3.5 Existing form of β-SiC

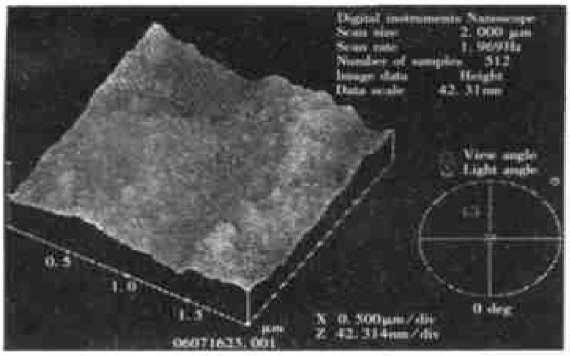

Fig. 14 AFM: Existing form of β-SiC

Fig. 14 is a zooming view of Fig. 13. Due to high hardness, β-SiC is left after polishing process. When Si infiltrates the green body, carbon reacts with Si to form β-SiC. So β-SiC exists in the surface of α-SiC or between the α-SiC grains, where are the very place covered or combined by the binder-the carbon-rich resin. The existing of β-SiC guarantees the well bonding between Si and α-SiC grains.

4 Conclusion

1. The ultrasonic treatment can improve the density of RBSiC.

2. During Si infiltrating, β-SiC is formed by the reaction of Si and carbon, which makes a well bonding between Si and α-SiC.

3. The RBSiC we fabricated possesses good polishability, and can be well polished by the float polishing.

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