IOP Conf. Series: Materials Science and Engineering **47** (2013) 012060 doi:10.1088/1757-899X/47/1/012060

The Effect of Si contents on the reaction-bonded Si_3N_4/SiC composite ceramics

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Abstract. Effect of Si contents on reaction-bonded Si_3N_4/SiC composite ceramics under pressureless was investigated. Si_3N_4/SiC composite ceramics were sintered at 1600℃ under nitrogen atmosphere by using SiC powders (1.5μm), Si powders (74μm) with different contents 37~55wt% and sintering additives Y_2O_3 as raw materials. The phases, microstructure and mechanical property were characterized by XRD, SEM, and compressive strength tests. The results demonstrated that when the content of Si powders was 37wt%, the more dense samples with the bulk density of 2.41 g/cm^3 and the higher compressive strength of 319 MPa could be obtained under pressureless.

1. Introduction

Si_3N_4/SiC ceramic as outstanding refractory could be used at high temperature 1500℃ in different atmospheres, because of its many properties such as high temperature strength, low thermal conductivity, thermal shock resistance, low coefficient of thermal expansion, chemical stability, excellent creep resistance and oxidation resistance. For the properties improvement of Si_3N_4/SiC ceramic, a new design concept (nanocomposite ceramic) was first introduced by K. Niihara and colleagues describing structural synergisms between the matrix Si_3N_4 and the nano-SiC in 1991 [1]. And the abundant studies demonstrated that high strength Si_3N_4/SiC nanocomposites were obtained by adding nano SiC powders to the Si_3N_4 matrix at different conditions, for example, hot-pressed sintering at high temperature 1850℃, sinter-post-HIP at about 2000℃, SPS method, et al[2-8]. So the preparation of Si_3N_4/SiC nanocomposites was costly and more requirements. While reaction-bonded Si_3N_4/SiC produced by reaction bonding of silicon powder compacts is economical compared to sintered Si_3N_4 and SiC because Si powder is much cheaper than that of Si_3N_4. Reaction bonding by direct nitridation of silicon powder at temperatures below its melting point (1414℃) is an attractive method to synthesize Si_3N_4/SiC composites. It has been reported that conventional Si_3N_4/SiC micron composite refractory was mainly prepared at about 1450℃ under pressureless sintering using larger micron particle size SiC powders, which compressive strength was below 300 MPa [9-13].

In the paper, the SiC fine powders 1.5μm was used as raw materials. The effect of different Si powders (74μm) contents 37~55wt% on phase, microstructure and compressive strength of reaction-bonded Si_3N_4/SiC composite ceramics under pressureless was studied and the content was optimized.

2. Experimental procedures

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Commercial 1.5μm SiC powders was used as the matrix phase and 74μm Si powders were used as associated phase in this study. Y_2O_3 (purity>99.9%, grain size is about 6μm) was used as sintering additives. Different content SiC powders and Si powders were first mixed by wet ball mixing with ethanol media in a plastic jar for 24h together with 5wt%Y_2O_3 (as sintering additives), and then dried to make a homogeneous powder mixture. PEG as binder was added to the powder mixture and mixed for 20 min. The resultant mixtures were put into a steel die to form compacts under certain pressure. Then the compacts were cold isostatically pressed under 200 MPa. At last, the compacts were nitrided at 1350℃ for 1 h and sintered at the temperatures 1600℃ for 3 h under nitrogen atmosphere.

The bulk densities of the sintered samples were measured by Archimedes method. Phase identification of the samples was performed by X-ray diffraction (XRD, Philips, X'pert Pro MPD). The microstructure of sintered samples was characterized by scanning electron microscopy (SEM, FEI, Nova 400 Nano). Compressive strength was obtained by dividing the peak load by the cross-sectional area of the sample.

3. Results and discussion

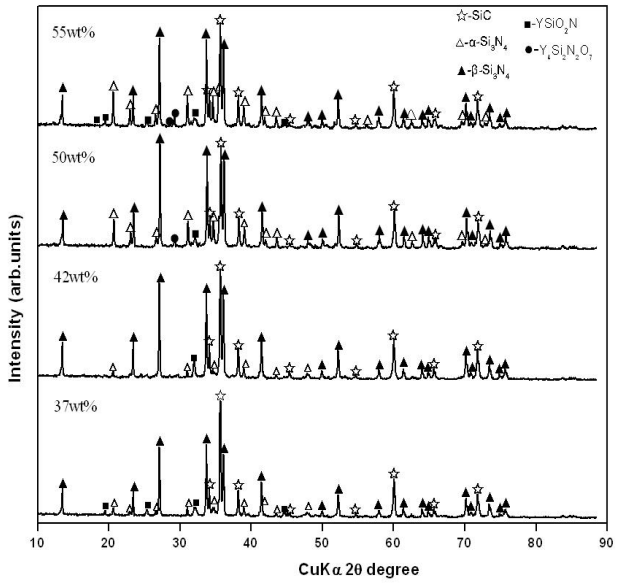

Figure 1. XRD patterns of samples with different Si contents

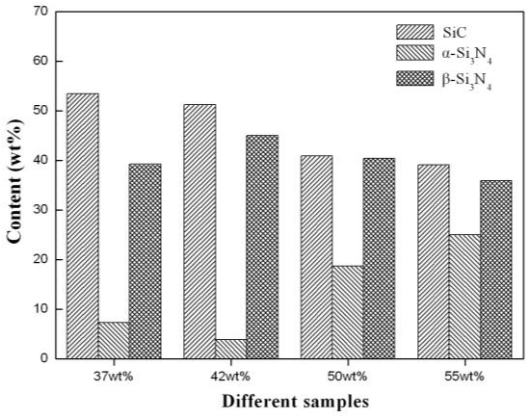

Figure 2. Different phase contents of samples with different Si contents

Figure 1 shows XRD patterns of samples with different Si contents. It could be observed that the major phases consisted of SiC, α-Si_3N_4 and β-Si_3N_4. There was a small amount of grain boundary phases and they were $YSiO_2N$ and $Y_4Si_2N_2O_7$. The location and intensity of SiC phase peak were not obviously changed in all samples. While the amount and intensity of α and β-Si_3N_4 phase peak were different in the four samples.

K (RIR) value method was introduced to do semiquantitative calculation of different major phases by the software JADE of XRD and the calculation result was showed in Figure 2. The content of SiC phase was decreased with the increasing of Si content. The content of α-Si_3N_4 was firstly decreased and increased, while β-Si_3N_4 was increased and then decreased. When the content of Si was 42wt%, α-Si_3N_4 was less and β-Si_3N_4 was more.

Figure 3 shows bulk density and compressive strength of samples with different Si contents. The bulk density and compressive strength were decreased with the increasing of Si content from 37wt% to 55wt% as shown in Fig.3. When the content of Si was 37wt%, the bulk density and compressive strength were higher and they were separately 2.41 g/cm^3 and 319MPa.

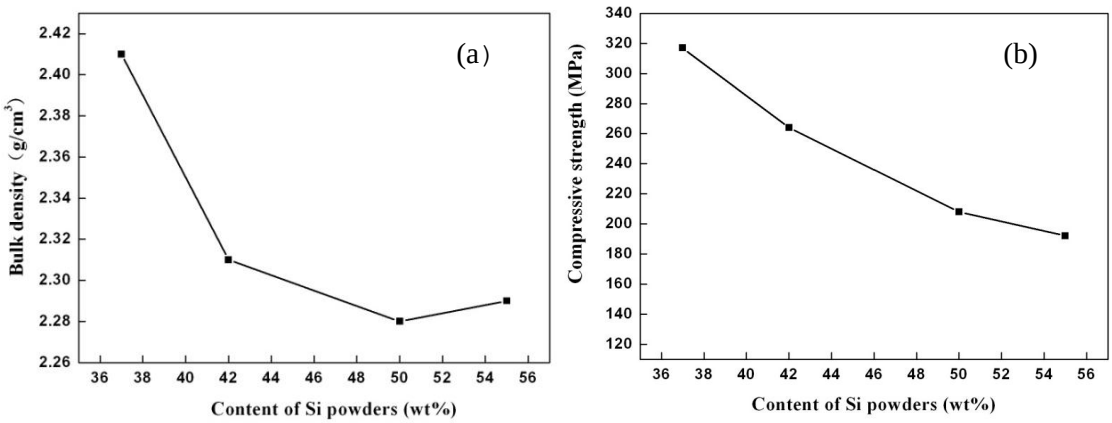

Figure 3. Bulk density (a) and compressive strength (b) of samples with different Si contents

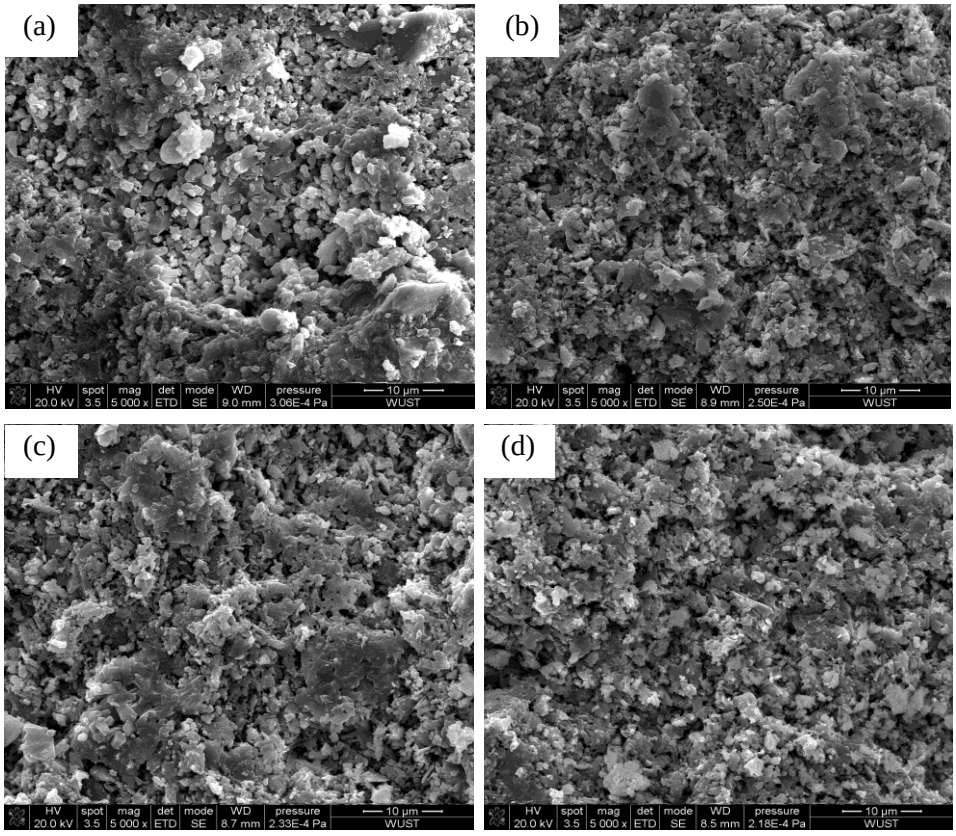

Figure 4. Microstructure photographs of samples with different Si contents: (a)37wt%; (b)42wt%;(c) 50wt%; (d)55wt%

Figure 3 shows the microstructure photographs of samples with different Si content: (a)37wt%; (b)42wt%;(c) 50wt%; (d)55wt%. It could be observed that the pore of (a) was less than the other three samples, which was consistent with the result of bulk density. When the content of Si was 37wt%, the combination among grains was compact and the grain crystallization was good. The rod-like grain was larger as shown in Figure 4. And the compressive strength was higher in Figure 3. There exist many small rod-like β-Si_3N_4 grains which distributes uniformly in Figure 4.5(b). And in the same sample,

the content of β-Si_3N_4 phase was higher as shown in Figure 2. A large number of β-Si_3N_4 grains present intertexture growth and much pore generated, which was confirmed by bulk density in Figure 3(a). It could be observed in Figure 4.5(c) that some agglomerated grains appeared. It may be because agglomerated Si particles distributed non-uniformly and increased the molten Si viscosity which was hard to flow. And much agglomerated Si_3N_4 grains could not make SiC combine well. So when the content of Si was 55wt%, the combination among grains was looser as shown in Figure 4.6(d) and the bulk density and compressive strength were lower in Figure 3. In conclusion, the optimized Si content was 37wt% when the sintering temperature was 1600℃.

Conclusions

Effect of Si contents on reaction-bonded Si_3N_4/SiC composite ceramics under pressureless was investigated. Si_3N_4/SiC composite ceramics were sintered at 1600℃ under nitrogen atmosphere by using SiC powders (1.5μm), Si powders (74μm) with different contents 37~55wt% and sintering additives Y_2O_3 as raw materials. The results demonstrated that the bulk density and compressive strength were decreased with the increasing of Si content from 37wt% to 55wt%. When the content of Si powders was 37wt%, the more dense samples with the bulk density of 2.41 g/cm^3 and the higher compressive strength of 319 MPa could be obtained under pressureless.

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