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Microstructure and mechanical properties of SiC platelet reinforced $BaOAl_2O_32SiO_2$ (BAS) composites

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Abstract

$BaOAl_2O_32SiO_2$ (BAS) glass–ceramic powders were prepared by sol–gel technique. SiC platelet reinforced BAS glass–ceramic matrix composites with high density and uniform microstructure were fabricated by hot-pressing. The effect of additional crystalline seeds on hexagonal to monoclinic phase transformation of Barium aluminosilicate was studied. The effects of SiC platelet content on the microstructure and mechanical properties of the composites were also investigated. The results showed that the flexural strength and fracture toughness of the BAS glass–ceramic matrix composites can be effectively improved by the addition of silicon carbide platelets. The main toughening mechanism was crack deflection, platelets' pull-out and bridging. The increased value of flexural strength is contributed to the load transition from the matrix to SiC platelets.

Keywords: B. Microstructure; C. Mechanical properties; D. SiC; Reinforced BAS composites

1. Introduction

One of the primary objectives of the NASA'S High Temperature Engine Materials Program(HITEMP) is to develop ceramic–matrix composites used for the hot section components in advanced aeropropulsion systems. Glass and glass–ceramics with low elastic modulus allow true reinforcement with high modulus ceramic materials such as short fibers, whiskers and platelets. $BaAl_2Si_2O_8$ is the only stable ternary compound in the ternary system $BaO–Al_2O_3–SiO_2$ [1]. Celsian $BaAl_2Si_2O_8$ (BAS), which has several advantages for being used as an ideal matrix material such as highly refractory nature, high melting temperature of 1760°C, low thermal expansion of 2.29×10^{-6}/°C up to 1000°C, excellent phase stability up to 1590°C, and good oxidation resistance [2,3]. In addition, the chemical compatibility between celsian matrix and silicon carbide make this material more attractive for being used as a matrix material of composites reinforced with silicon carbide for thermostructural applications. Ceramic matrix composite was fabricated by adding a second phase, such as whiskers or platelets to get high toughness and strength. The toughening and strengthening mechanisms of SiC whiskers/platelets are effective at elevated temperature. However, it is difficult to achieve uniform dispersion of whiskers, especially in a high volume content, and the potential health hazards in processing encourage man directing attention to other forms of particulate reinforcement, in particular, single-crystal platelets. The further cause is that platelets can give better fracture resistance than whiskers although they may reduce the strength.

To date, however, the reports on BAS composites are limited. Most of the research work has focused on how to prepare BAS powder and promote transformation from hexagonal phase to monoclinic phase of BAS. It was found that hexagonal phase is the first crystallization phase of the melt. This phase is metastable below 1590° and will undergo a reversible transformation to an orthorhombic crystal structure at 300°C, accompanying a volume change in the range of 3–4 vol.% increment. This phenomena makes it undesirable as structural material [4]. The method for perfectly promoting transformation from H→M has not been reported. Our objective is to prepare SiC platelets reinforced BAS composites

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with high density and uniform microstructure, to get a preferred monoclinic phase of BAS in the composites.

2. Experimental procedures

2.1. BAS powder preparation

The stoichiometry BAS powder with compositions of BaO 40.8 wt.% Al_2O_3 27.2 wt.%, $SiO_2$32 wt.% was fabricated by Sol–gel technique using TEOS, Aluminum isoproxide, Barium acetate as source of silicon, aluminum and barium. The procedure is described in Fig. 1.

Aluminum alkoxide and TEOS have a good solubility in alcohol organic solvent [5]. So we can mix aluminum alkoxide with TEOS at molecular level in alcohol without water. Acetate acid was added into the mixture for the purpose of preventing the precipitation of metal hydroxides by slowing down the hydrolysis and condensation rate of aluminum alkoxide, and catalysis hydrolysis of TEOS. We called it solution A. Then the mixture, solution A, was poured into the solution of Ba-acetate in acetate acid, alcohol and water with vigorous stirring till the solution became clear. The ideal for this process depends on the quicker rate of hydrolysis and condensation of aluminum alkoxide to form the network. With the aging time going, dispersed molecular TEOS chemically incorporate in the network. This procedure involves two steps: the hydrolysis of metal alkoxide produces metal hydroxides, followed by polycondensation of hydroxyl groups to form a metal oxide network. The barium acetate molecule will disperse in the interstice of the network. So we can get BAS dried gel with a molecular level dispersion at low temperature by Sol–gel [6]. Then xerogel was heat-treated to remove all organic material.

2.2. Composite fabrication

The material used in this study was BAS glass–ceramic reinforced with SiC platelets. The content of reinforcement was changed in the range 0–40 vol.%.

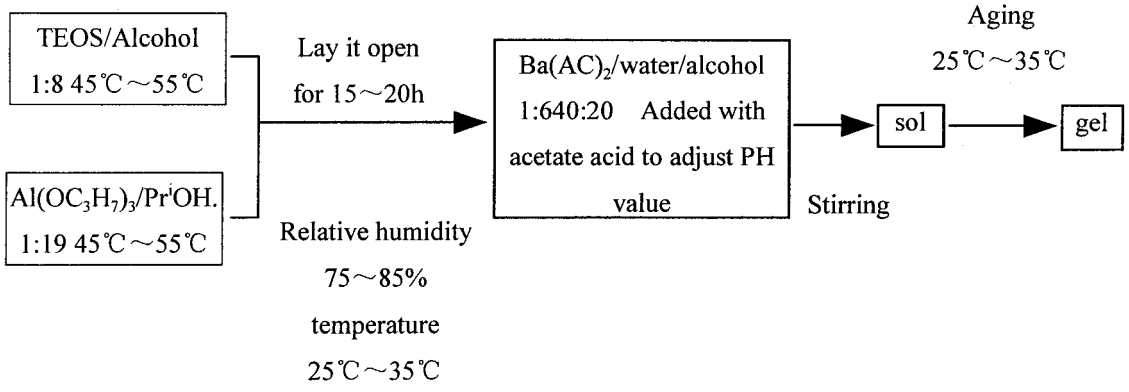

Fig. 1. Synthesis procedure of BAS gel.

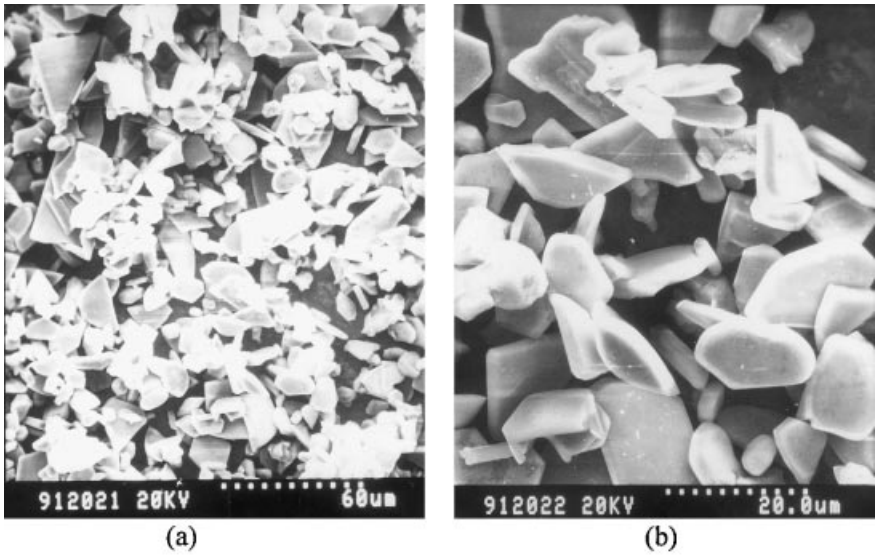

Fig. 2. SEM morphologies of as-received SiC platelets: (a) at low magnification; (b) at high magnification.

SiC platelets (C-Aris Technology, SF grade, made in Canada) with width and aspect ratio about 9 –24 and 4–12 μm respectively, as shown in Fig. 2.

The powders were mixed by ball milling in alcohol for 12 h to ensure a good dispersion. After milling, the balls were removed and the mixture was dried. Then the mixture were put into graphite die and hot-pressed according to procedure as following:

950°C, 4–5 Mpa, 0.5 h + 1370°C, 7–15 Mpa, 1 h

Composites were machined to the bar specimens for the single-edge notched-beam (SENB) with the

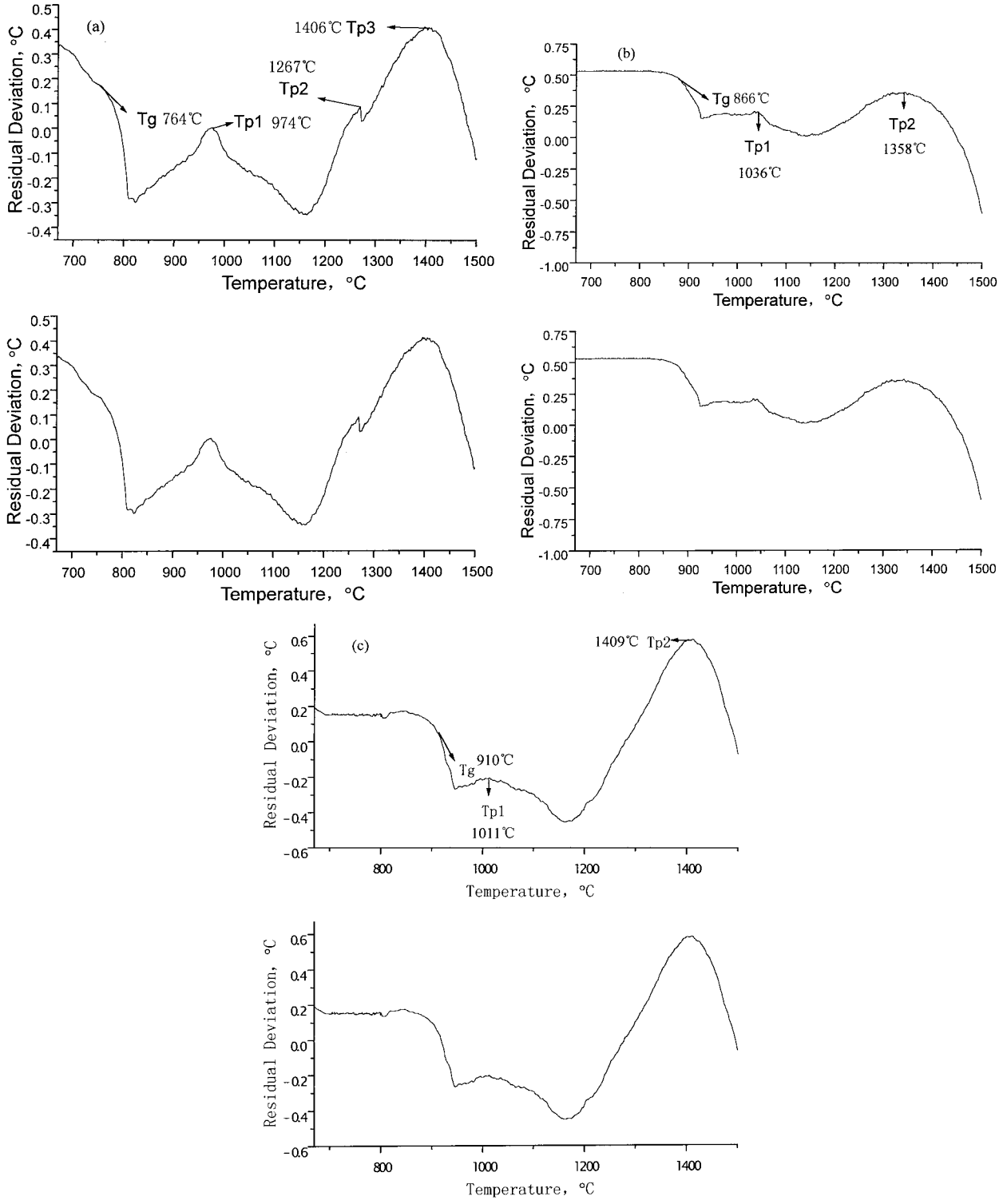

Fig. 3. Trace of DTA. (a). Lithium oxide was added to pure BAS. (b). Pure BAS. (c). 20 wt.% seed was added to pure BAS.

dimensions of 2×4×20 mm, and a notch of 2 mm in depth, and specimens for three-point bending to determine flexural strength with dimensions of 3×4×36 mm.

2.3. Characterization

The densities of composites were measured by Archimedes method. The flexural strength (σ_f) and fracture toughness (K_{IC}) were measured in air at room temperature using a Instron1186 testing machine. The flexural strength test was performed on the specimens with a span of 30 mm. K_{IC} was measured by SENB with a span of 16 mm. The cross-head speed was 0.5 mm/min for σ_f and 0.05 mm/min for K_{IC} measurement.

XRD(CuK_a) was used for phase analysis. Fracture surface and crack propagating paths produced by vickers indentation were examined using scanning electronic microscopy (SEM). The microstructure of composites characterized by transmission electron microscopy (TEM) and energy dispersive spectroscopy (EDS).

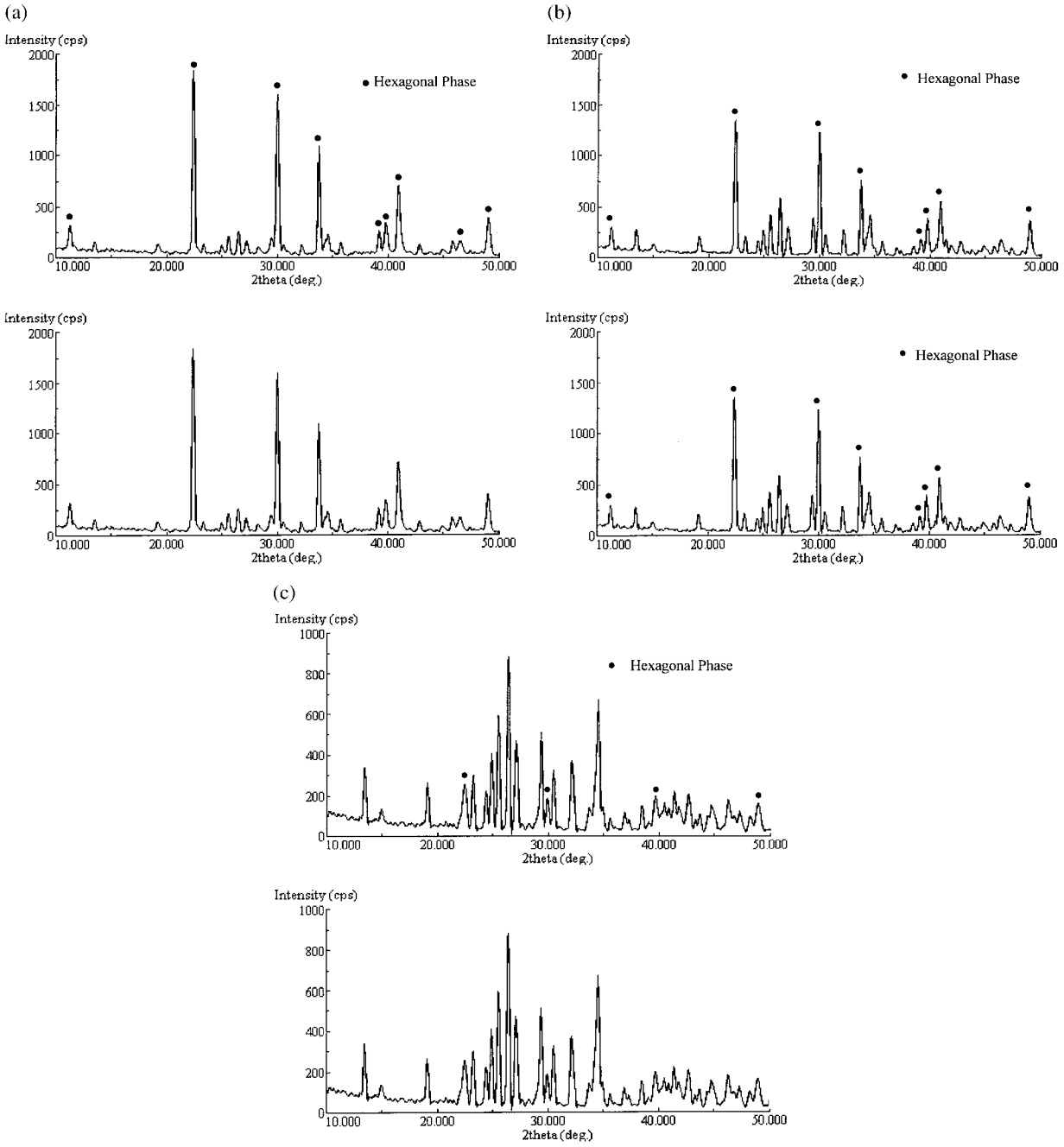

Fig. 4. XRD profiles of BAS. (a) BAS with added 5 wt.%seed heat-treatment at 1450°C for 24 h. (b) BAS added with 20 wt.% monoclinic phase at 1200°C for 1 h, (c) Lithium oxide added to BAS at 1200°C for 1 h.

3. Results and discussion

3.1. Crystallization of BAS glass

DTA traces of stoichiometric glass BAS are shown in Fig. 3. T_g of 20 wt.% seed added to pure BAS was determined to be 910°C, a value much higher than that of lithium oxide added to BAS glass for preparing seed to promote phase transformation from H→M of BAS. The peak temperatures of BAS, lithium oxide added to BAS glass, BAS seed are 1036,1358, 974,1267,1406, 1011,and 1409°C respectively.

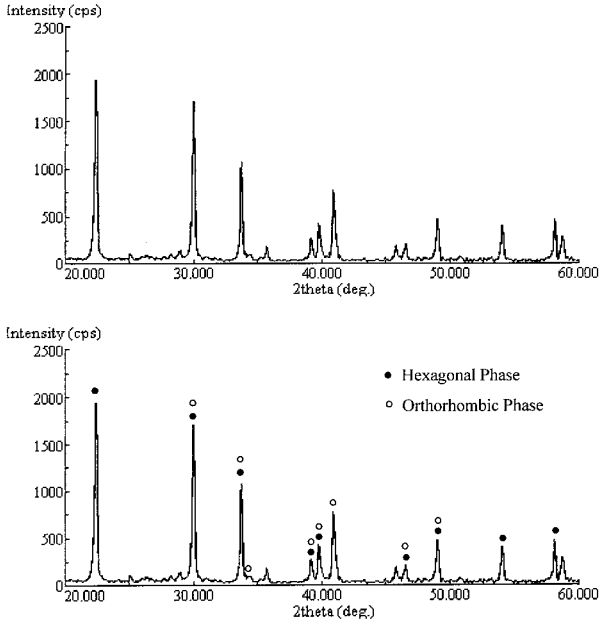

Fig. 5. XRD profile of BAS without seed of celsian. The crystalline phase was predominately hexagonal polymorphs.

The XRD results of BAS with or without seed are revealed in Figs. 4 and 5. The hexagonal phase was obtained rather than the preferred monoclinic phase in BAS glass. Only a limited amount of celsian formed by delaying the time of heat-treatment. But BAS glass added with lithium oxide contained almost, the monoclinic phase. BAS added seed the monoclinic phase of BAS was predominant. The crystallization of monoclinic celsian could be largely enhanced by the addition of seed, because it provides a nuclear for celsian. Lithium oxide lowered the glass transition temperature of the immediately adjacent gel matrix and led to the crystallization of pure monoclinic celsian. The stabilization effect of Li_2O on celsian crystallization in BAS glass was attributed to a reduced viscosity in the presence of Li_2O. As shown in DTA trance, adding Li_2O lowered T_g. The monoclinic phase could nucleate directly in the bulk, so monoclinic phase could be achieved meanwhile preventing hexagonal phase formation. The seed acted as normal heterogeneous centres and epitaxial substrates for monoclinic celsian and produced pure monoclinic celsian in some temperature regions.

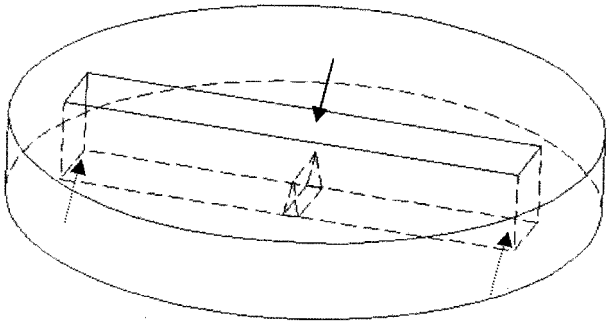

Fig. 7. Schedule drawing for specimens machined and tested.

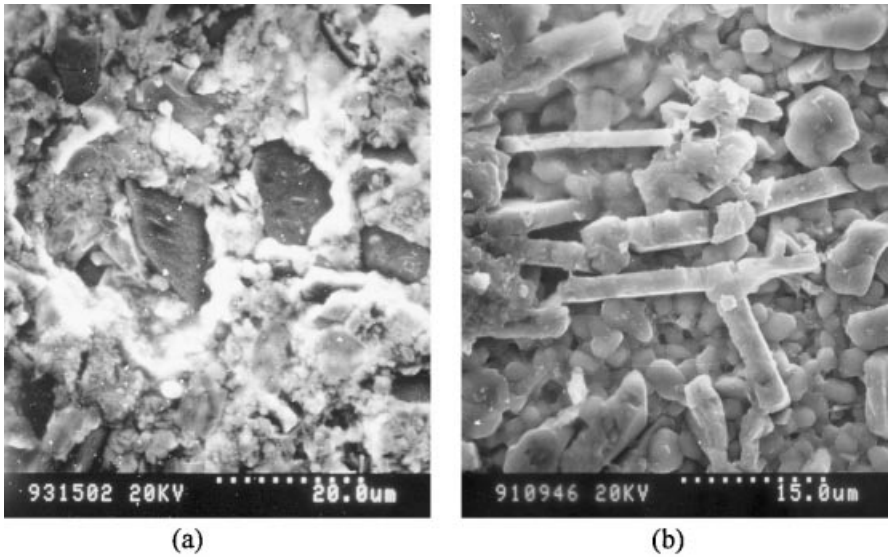

Fig. 6. SEM morphologies for polished surface of the composite with content 20 vol.% SiC_{pl}. (a) Perpendicular to hot pressing direction. (b) Parallel to hot pressing direction.

3.2. Microstructure of the composites

During the hot pressing the broad surface of the SiC platelets in the composites oriented preferentially along the plane of hot pressing, as shown in Fig. 6. The broad surface of the platelets preferential alignment along hot pressing plane and the thickness surface of the platelets along the hot-pressing direction were observed from the SEM morphology of the polished surface. The bar specimens were machined as shown in Fig. 7. During three point bending tests (σ_f and K_{IC}), crack propagating direction is perpendicular to the hot pressing plane, so that the SiC platelets could stop more cracks propagation which results in higher values of σ_f and K_{IC}.

The TEM morphologies of the composite with content of 30 vol.% SiC_{pl} are shown in Fig. 8. The silicon carbide platelets are directly bonded to the matrix glass. This microstructure will benefit for load transition.

Fig. 9 shows XRD result of the composite with content of 40 vol.%. From Fig. 9, we can see that the phase of the SiCpl/BAS composite is consisted of celsian and α-SiC with a few of hexacelsian. Celsian seeds can effectively promote the H→M BAS transformation in the composite, as shown in Fig. 9. The choice of reinforcement is limited by high temperature chemical incompatibility and by thermal expansion mismatch with the matrix. The preferred monoclinic phase could be obtained

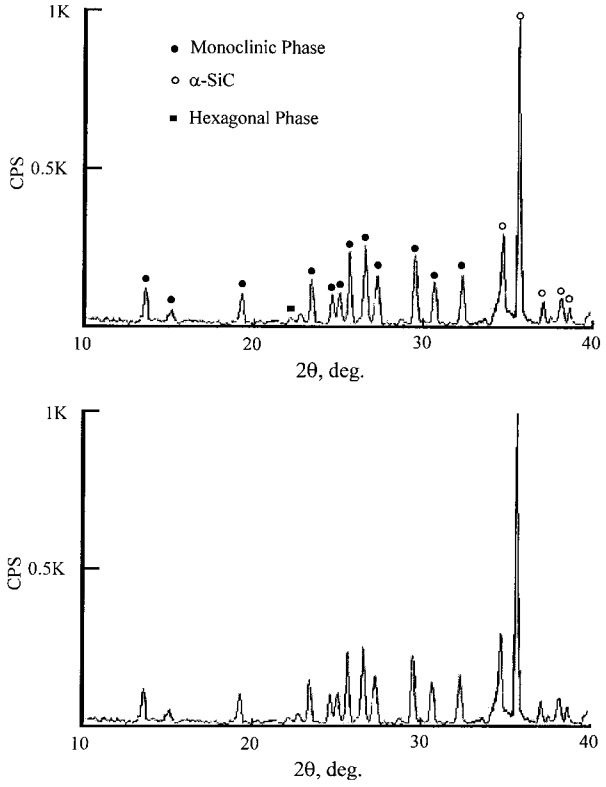

Fig. 9. XRD profile of BAS added with 40 vol.% silicon carbide platelets.

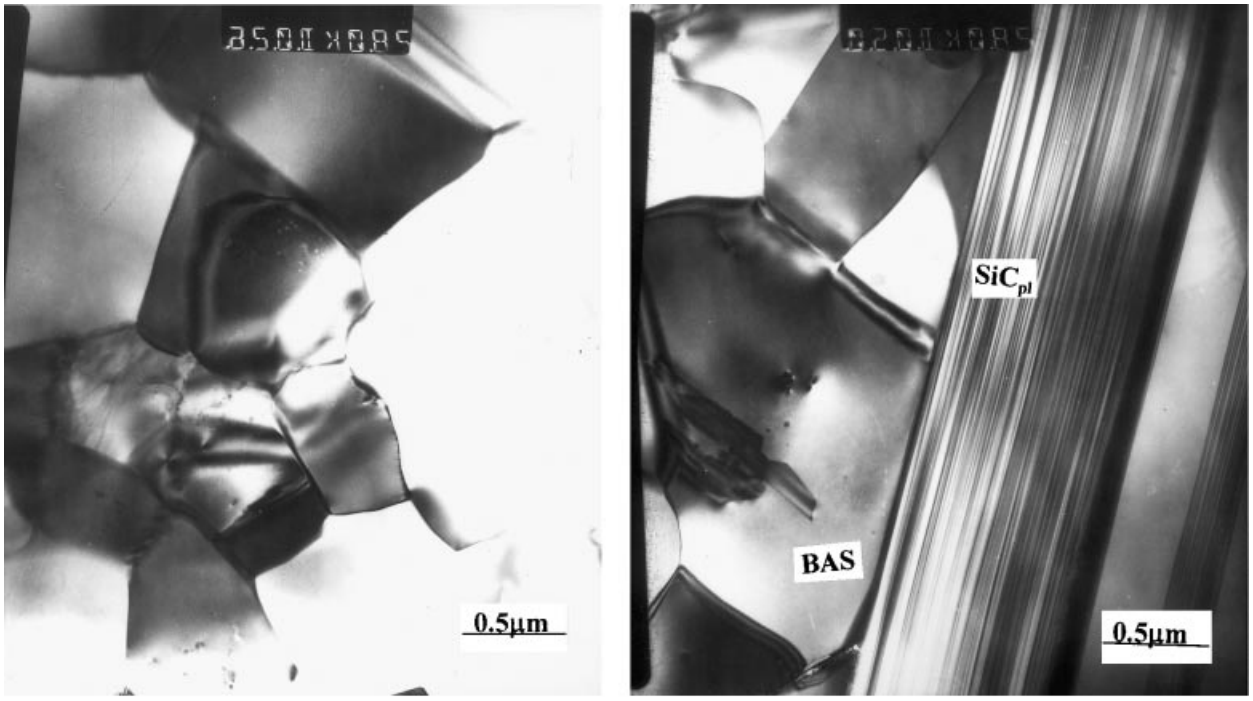

Fig. 8. Bright field morphologies of TEM for SiCpl/BAS composite with content of 30 vol.% SiC_{pl}. (a) Matrix grains bond to each other. (b) SiC platelets bond to matrix grains.

in the composites by adding celsian seeds. The thermal expansion coefficient for celsian is reported to be little less to that for silicon carbide whiskers. The thermal expansion coefficient for silicon carbide platelets is $3–3.5\times10^{-6}/°C$, a bit more than thermal expansion coefficient for celsian. So thermal residual stress between the matrix and reinforcement can be reduced. Elastic modulus of silicon carbide platelet reinforcement is greater than that of matrix, therefore, this benefits to physical compatibility between matrix and reinforcement. Interface bonding strength between reinforcement and matrix of BAS provides suitable conditions for load transition effect. So it benefits by enhancing mechanical properties of composite.

3.3. Mechanical properties

The mechanical properties of the composites reinforced with silicon carbide platelets are shown in Figs. 10 and 11. The flexural strength and fracture toughness increase with increasing platelet content in the range of 0–30 vol.%,then the flexural strength and fracture toughness decrease slightly with further increasing content of silicon carbide platelets. Due to thermal expansion coefficient between the reinforcement and BAS matrix, by adding 30 vol.% silicon carbide platelets, the fracture toughness and the flexural strength are improved 2.13, 1.82 times compared with properties of pure BAS from the Fig. 11. Silicon carbide platelets were confirmed more effective in enhancing fracture toughness than flexural strength (the cause was given below).

SEM fracture surface morphologies and crack path generated by indentation are shown in Figs. 12 and 13. It can be seen that the platelets pull-out and cavity could be observed in the roughness fracture surface. In Fig. 13, the grain of BAS and reinforcement interaction zone behind the crack tip could be seen. In fact, it is believed that this interaction effect between the propagating crack and microstructure leads to the high K_{IC}

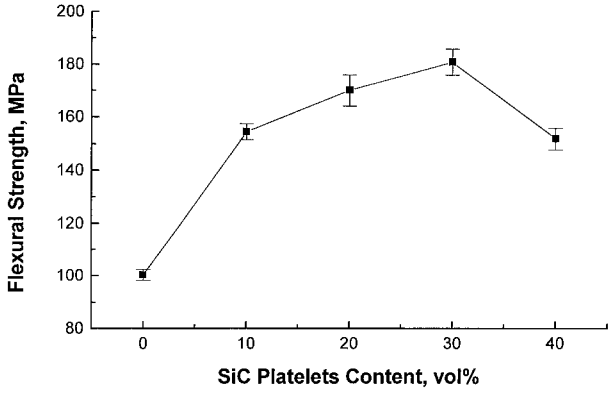

Fig. 10. Flexural strength of the composites as the function of SiC platelet.

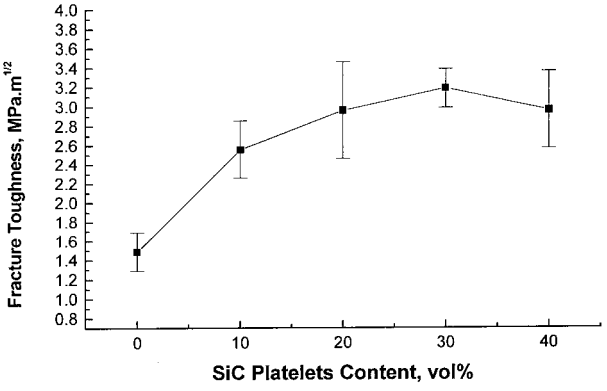

Fig. 11. Fracture toughness of the composites as the function of SiC platelet.

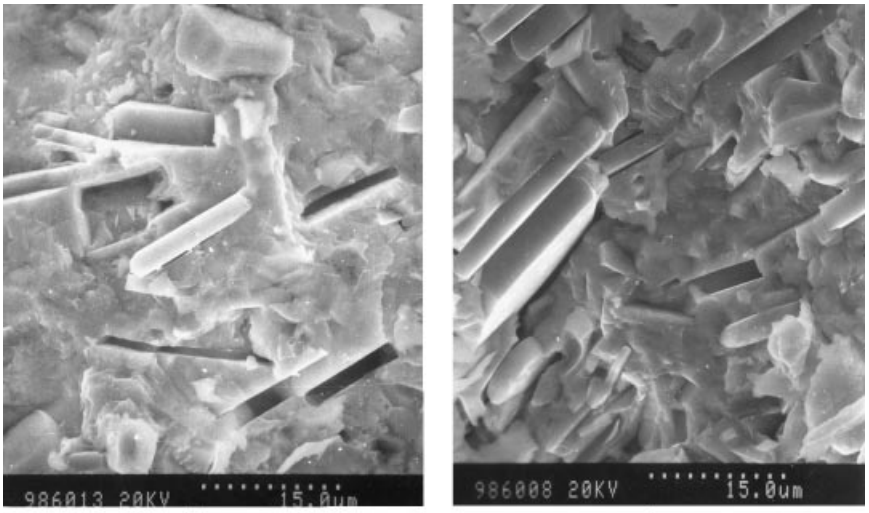

Fig. 12. SEM fracture surface of the composites. (a) BAS with 30 vol.% SiC_{pl}. (b) BAS with 40 vol.% SiC_{pl}.

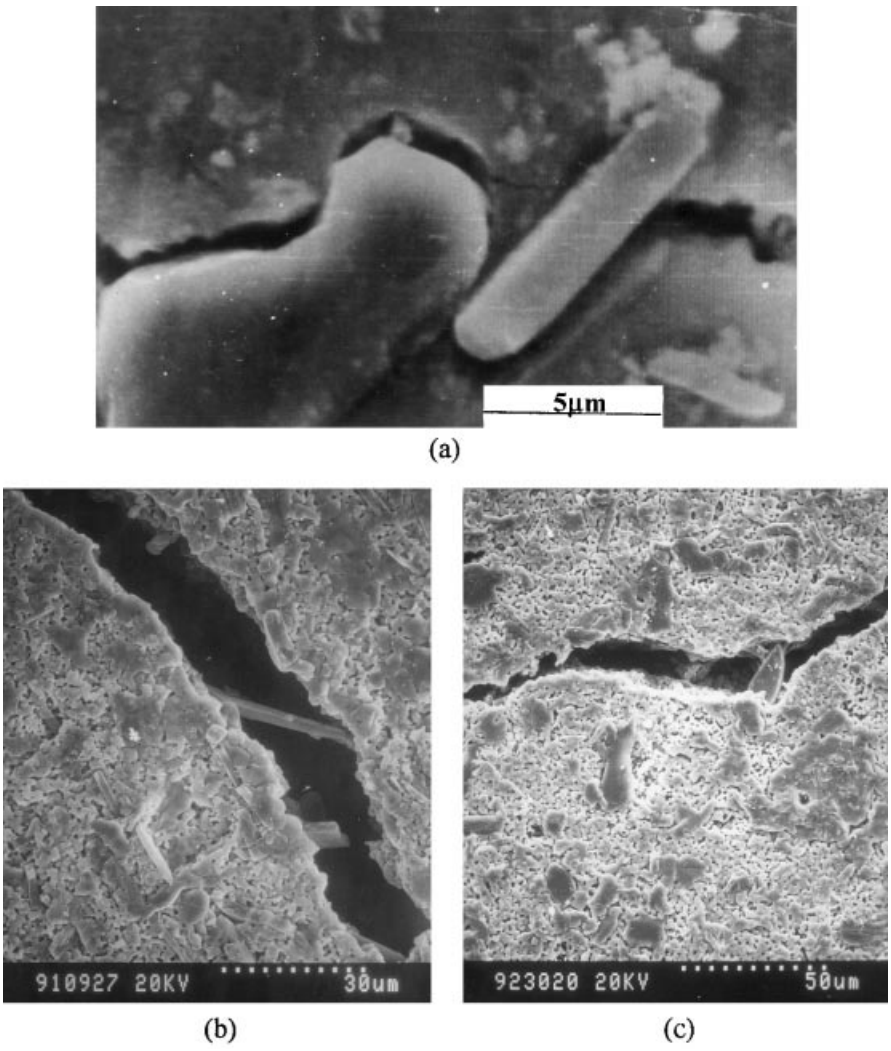

Fig.13. SEM morphologies of indentation crack path of the composites with content of 20 vol.% SiC_{pl}. (a) Crack deflection; (b) crack bridge; (c) crack pull-out.

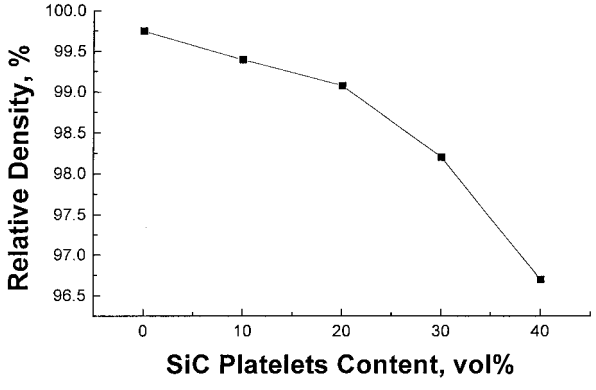

Fig. 14. Relative density of the composites as the function of silicon carbide platelets.

value. The extensive crack interactions with the platelets such as crack deflection, platelet bridging and pull-out, are observed more clearly by the crack propagating paths produced by vickers indentation. It reveals the strong interaction in the crack morphology. The interaction can devalue the intensity of crack tip, that is, SiC platelets were effective in improving the crack propagating resistance.

Increasing content of SiC platelet in the composites, the relative density of composites is slightly decreased as shown in Fig. 14. All densities of the composites are more than 96%. Although strength and fracture toughness are sensitive to relative density, but in this paper, fracture toughness and flexural strength are decreased with further increasing content of SiC platelets as the volume fraction of SiC greater than 30% should have no relation to the relative density.

4. Conclusions

The following conclusions from the results of this investigation can be drawn:

1. Amorphous stoichometric glass BAS powder could be obtained by Sol–gel technique using metal alkoxide and inorganic material barium acetate.
2. Hexagonal phase was obtained rather than the preferred monoclinic phase in BAS glass. But by incorporation of 3 wt.% LiO_2 to BAS can effectively promote hexagonal phase of BAS to celsian, and the result show that using it as monoclinic seed also could promote transformation from hexagonal to monoclinic.
3. By adding seeds of monoclinic BAS, the phase of SiCpl/BAS composites is constituted of celsian, hexagonal phase of BAS and α-SiC. It benefits to enhance mechanical properties of SiCpl/BAS composites.
4. BAS matrix composites reinforced with SiC platelets with uniform microstructure and more than 96% density of theoretic could be fabricated by hot pressing method.
5. The flexural strength and fracture toughness increase with increasing platelet content in the range of 0–30 vol.%,then the flexural strength and fracture toughness decrease slightly with further increasing the content of silicon carbide platelets.

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