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ARTICLE

Liquid Phase Flow Behavior and Densification Mechanism of Al/B_4C Composites Fabricated via Semisolid Hot Isostatic Pressing

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Abstract: Aluminum matrix boron carbide (Al/B_4C) composites are important thermal neutron shielding materials. In order to prepare B_4C/6061Al composites with higher density, Al/B_4C composites reinforced by 30 wt% B_4C particle were fabricated via hot isostatic pressing (HIP) in the semi-solid temperature range of 6061Al. Microstructures, mechanical properties and densification mechanism of the composites were investigated. The results indicate that the density of Al/B_4C composites fabricated by the semisolid hot isostatic pressing is very close to the theoretical density. Although the tensile strength can reach up to 300 MPa, the ductility of the semisolid HIP Al/B_4C composites is not good mainly because of the high content of B_4C. Aluminum liquid phase is observed indirectly which mainly forms during semisolid HIP process, and it is helpful to improve the combination between the B_4C particles and the 6061Al matrix. The increase of density and strength property is also mainly due to the effect of aluminum liquid flowing into the internal gaps under the hot isostatic pressing condition with high temperature and high pressure.

Key words: Al/B_4C composites; semisolid; liquid phase; densification mechanism

Boron carbide (B_4C) has low density, high strength, high specific stiffness, good damping capacity and excellent thermal conductivity[1]. B_4C is a proper candidate as reinforcement material in Al matrix composites. Recently, aluminum matrix boron carbide composites (Al/B_4C) have been widely used in the fields of aviation industry, cycling industry, electronic communication[2,3] etc. For containing the isotope of ^{10}B, the thermal neutron shielding properties of the Al/B_4C composites have also been concerned[4]. The Al/B_4C composites are usually prepared by methods of powder metallurgy (PM) or melting[5-8]. However, because of the weak wettability of Al and B_4C at low temperatures, the densities of the Al/B_4C composites fabricated by PM are quite low, which results in poor mechanical properties. For the melting method, it is also difficult to achieve even dispersion of B_4C particles in the Al matrix. Therefore, conventional powder metallurgy or melting is not an effective way for preparing Al/B_4C composites with enhanced mechanical and neutron shielding properties[9-11].

The technique of semisolid processing, developed in 1970s, can process alloys in the states of solid and liquid[12]. Semisolid processing has several advantages in fabricating composites, and the matrix will show low deformation resistance and good formability, which results in significant improvement in density of alloys[13,14]. Besides, because the semisolid processing temperature is much lower than the casting temperature, the deleterious interfacial reaction between reinforcing particle phase and matrix phase will be effectively restrained[15-17].

Hot isostatic pressing (HIP) is a sintering technique containing hot pressing and isostatic pressing, which usually processes alloys in the state of solid. Materials fabricated using HIP usually show improved microstructures and mechanical properties[18,19]. However, there are few reports on

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the combination of semisolid processing and HIP. Even with relevant research [20,21], they did not make deep association between semisolid technology and HIP technology in process and theory. In order to improve the microstructures and mechanical properties, the semisolid HIP process was used to prepare Al/B_4C composites in this research, and we attempted to explain the densification mechanism of the semisolid HIP on composites. Al/B_4C composites were also prepared by conventional vacuum sintering (CVS) at the same temperature for comparison.

1 Experiment

1.1 Materials

Raw materials were commercial powders of 6061Al and B_4C. 6061Al powders were fabricated by gas-atomizing 6061 aluminum alloy with a near-spherical morphology (mean particle size was 40 μm), whose chemical composition is listed in Table 1. B_4C particles were prepared by carbon thermal reduction with a mean particle size of 10 μm, whose chemical composition is summarized in Table 2.

1.2 Prepare process and thermal analysis

30 wt% B_4C particles and 70 wt% 6061Al powders were mixed with a certain percentage of steel balls and milling media. After milling for 4 h, the milled powders were dried in an evacuated container. The dried powders were cold isostatically pressed into compacts. After sealed in steel packages, the compacts were hot isostatically pressed at a pressure of 30 MPa and a temperature of 650 °C for 0.5 h. For comparison, compacts prepared by cold isostatic pressing (CIP) were also sintered in a vacuum (5×10^{-3} Pa) at a temperature of 650 °C for 0.5 h.

To measure the liquid volume fraction, a sample of the 6061 aluminum alloy powder (30 mg) was placed into an alumina pan and then heated from room temperature to 700 °C at a rate of 10 °C/min using Netzsch-STA 449C deferential scanning calorimeter (DSC) equipment under argon gas flow of 20 mL/min, and the DSC curve is shown in Fig.1a. It indicates that the actual freezing range of 6061 aluminum alloy powders is 614.4~673.9 °C, which is the basis for selecting the semisolid hot isostatic pressing temperature. The solid fraction and liquid fraction versus temperature curve was obtained by integration of the DSC curve (Fig.1b). The liquid volume fraction of the samples at 650 °C is 15 vol%. The semisolid treatment window is approximately 20 °C, as shown in Fig.1b, with the liquid fraction ranging from 5 vol% (at 640 °C) to 35 vol% (at 660 °C).

Table 1 Chemical composition of 6061Al powder (wt%)

Mg	Si	Cu	Cr	Fe	Al
0.92	0.57	0.21	0.12	0.09	Bal.

Table 2 Chemical composition of B_4C particle (wt%)

B_2O_3	Fe_2O_3	B	C	B_4C
0.50	0.49	0.48	3.68	Bal.

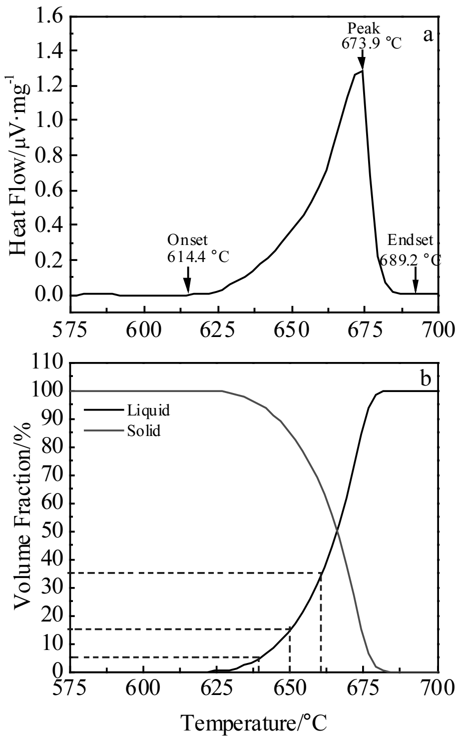

Fig.1 DSC curve of 6061 Al alloy powders at a heating rate of 10 °C/min (a) and volume fraction of solid and liquid versus temperature derived from the DSC curves (b)

1.3 Test method

The mechanical properties were investigated using a WANCE-100 mechanical testing machine. The densities of the Al/B_4C composites were tested by Archimedes method. Microstructures and fracture morphologies of the Al/B_4C composites, and interface between composites and steel packages were observed by scanning electron microscope (SEM, FEI SIRION200). The component elements were analyzed using energy dispersive spectroscopy (EDS).

2 Results and Discussion

2.1 Microstructure

Fig.2 shows microstructures of Al/B_4C composites fabricated by vacuum sintering and semisolid hot isostatic pressing. As indicated by arrows in Fig.2a, polygon B_4C particles are homogenously distributed in the Al matrix in vacuum sintered composites. However, many pores are observed between the B_4C particles and the Al matrix, which suggest a weak interface combination. Composites prepared using semisolid HIP also show a homogenous dispersion of B_4C particles in Al matrix (as shown in Fig.2b). However, no pores or apertures are observed between the B_4C particles and the Al matrix, indicating an enhanced combination. This enhancement is due to a better wetting property of Al liquid formed at the applied high pressure during the semisolid HIP process.

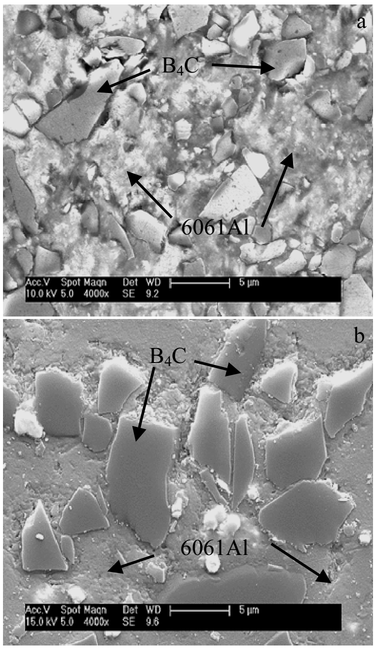

Fig.2 Microstructures of Al/B_4C composites prepared by conventional vacuum sintering (a) and semisolid HIP (b)

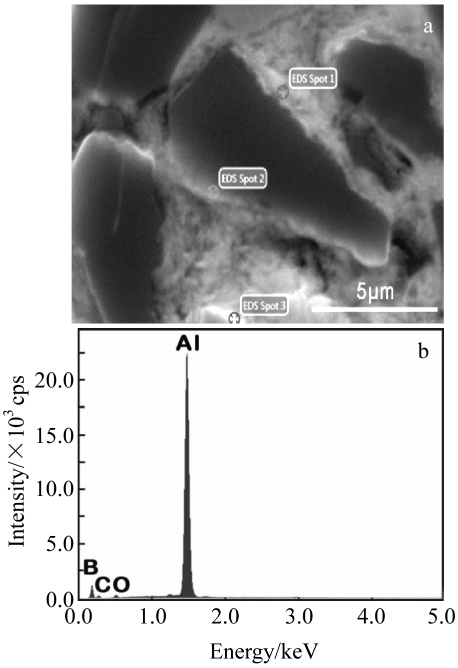

Fig.3 SEM image (a) and EDS spectrum (b) of Al/B_4C composites prepared by semisolid HIP

Detailed microstructure observation was conducted on the bonding interface between the B_4C particles and the Al matrix for composites prepared using semisolid HIP process. SEM image at high magnification and EDS results are shown in Fig.3. From the comparative analysis, it can be seen that the shapes of some B_4C particles turn into sub-orbicular after the semisolid HIP, which is somewhat different from the polygonal shape in Fig.2a. This change in shape suggests that 6061Al has a certain degree of chemical bonding with B_4C particles, which can bring some benefits for improving the interface bonding. EDS tests were also conducted on the bonding interface of Al-B_4C at three different places (as shown in Fig.3b), and the results indicate that the same peak of Al element appears at the surface of B_4C particles, suggesting that there is a strong coalescent between the B_4C particles and the Al matrix.

2.2 Surface morphology

To further certify the formation of the liquid phase of aluminum, the composites were stripped from the steel package after semisolid HIP and the outer surface was investigated using SEM and EDS (Fig.4). The results indicate that B_4C particles cannot be recognized in the outer surface, and instead the surface is covered with a uniform layer of Al matrix. Elements of Mg and Si are detected only on partial surface, which are all alloying elements of 6061 Al. It is suggested that element distribution on the surface is not uniform, and a portion of Al matrix melts are extruded into the interface between the composite and steel package during the process of semisolid HIP.

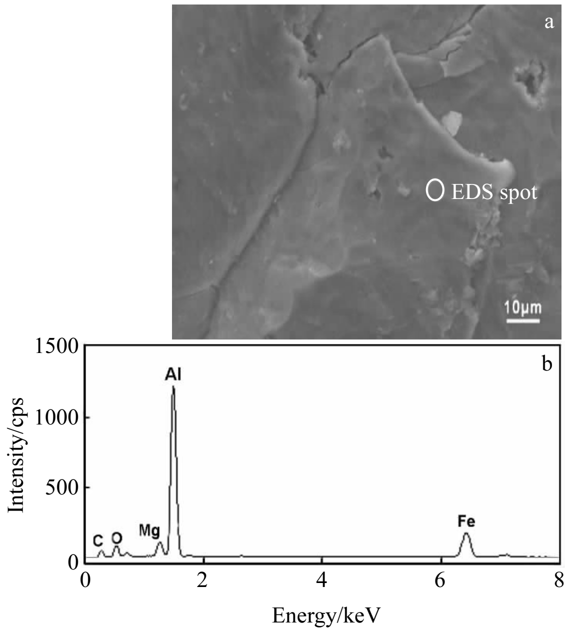

Fig.4 SEM morphology (a) and EDS analysis (b) of the surface of Al/B_4C composites prepared by semisolid HIP

2.3 Densification mechanism

The densities of Al/B_4C composites prepared by different processes are listed in Table 3. The density of conventional vacuum sintered compact is 91.9% of the theoretical density, while the density of the semisolid HIP compact is 99.6% of the

Table 3 Density of Al/B_4C composites prepared by different methods

Process	CIP	CVS	Semisolid HIP	Theory density
Density/g·cm^{-3}	2.02	2.42	2.66	2.67

theoretical density, suggesting that the compact cannot achieve a high density during the conventional vacuum sintering. Therefore, the density of composites prepared by semisolid HIP is significantly improved and close to theoretical density.

In order to clarify the mechanism of the densification process, a typical free Al/B_4C surface which is not covered by the steel package was further analyzed by SEM and EDS (Fig.5). The results indicate that many spherical fine particles exist on the free surface and they are mainly composed of Al. The main elements on the area without spherical particle are Al, Mg, Si, C, etc, which are consistent with the composition of the 6061Al. These two kinds of element distributions are consistent with the results of the interface between composite and steel package.

Based on the above observation, a densification mechanism is proposed, as shown in Fig.6. Under the condition of semisolid HIP process, the densification process of Al/B_4C composite can be mainly described by three steps. Firstly, when the temperature during HIP reaches the semisolid temperature range of 6061 Al alloy, some liquid phases between particles appear (Fig.6a). Secondly, at the high temperature and high pressure applied, the volume of the compact shrinks, and the Al liquid phase begins to flow and fills the internal pores or gaps in the compact (Fig.6b). Thirdly, after the composite compact is fully densified, the Al liquid overflows to the surface. Due to the weak surface tension and poor interfacial wettability, the Al liquid does not spread on the free surface, but appears in the form of spherical particles (Fig.6c). If the spherical liquid phase receives the constraint and extrusion from the cladding, it will fill the gap between the steel package and the composite material. This is the phenomenon shown in microscopic analysis of Fig.4a: after liquid flattening, the outer surface of the composite is covered.

2.4 Mechanical properties and fracture behavior

The tensile strength of the conventional vacuum sintering compact (86 MPa) is less than one third of that of the semisolid HIP one (301 MPa), indicating that semisolid HIP process can effectively improve the strength. The elongation of the two kinds of samples does not exceed 3%, and no obvious yield behavior is found in the two kinds of composites. The poor elongation suggests that doping a percent of 30 wt% B_4C results in a deterioration of ductility.

Fig.7a shows the fracture morphology of the Al/B_4C composites prepared by conventional vacuum sintering. Many fine B_4C particles are observed to be embodied in the Al matrix phase, indicating a good bonding between Al and fine B_4C particles. However, the combination between the Al matrix and the large particles are relatively weak, because there are a lot of clear holes. The existence of a large number of holes may be the main reason for the low strength and elongation of the vacuum sintered composites.

The fracture morphology of Al/B_4C composites prepared by semisolid HIP is shown in Fig.7b. Numerous dimples can be observed in the fracture morphology, suggesting a certain extent of plasticity. Although dimples are often considered as a sign of

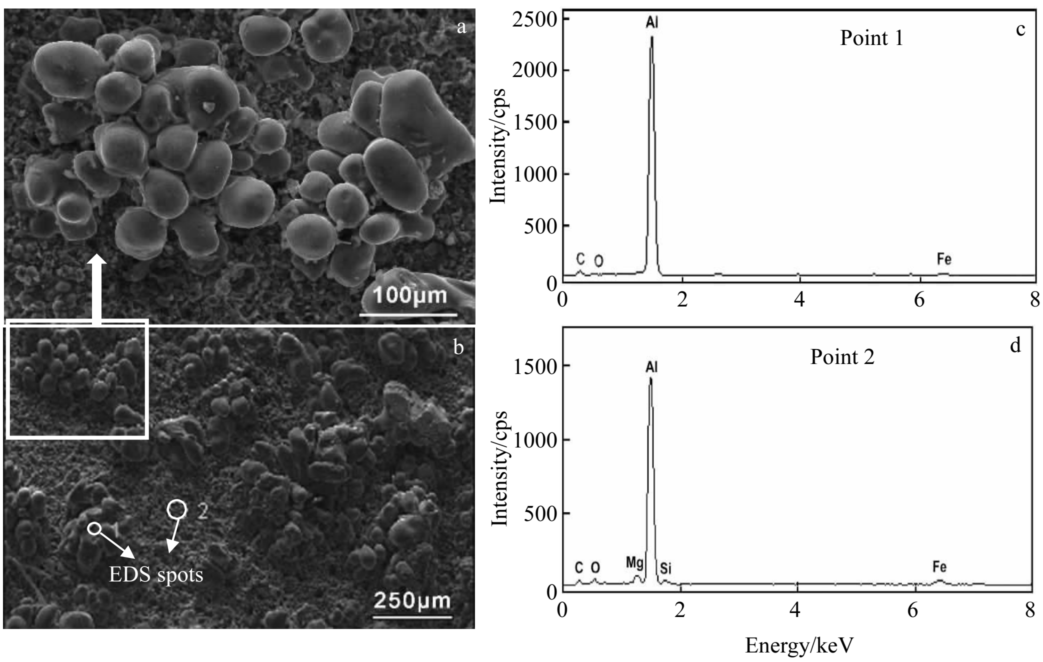

Fig.5 SEM images (a, b) and EDS spectra (c, d) of the free surface of Al/B_4C composites

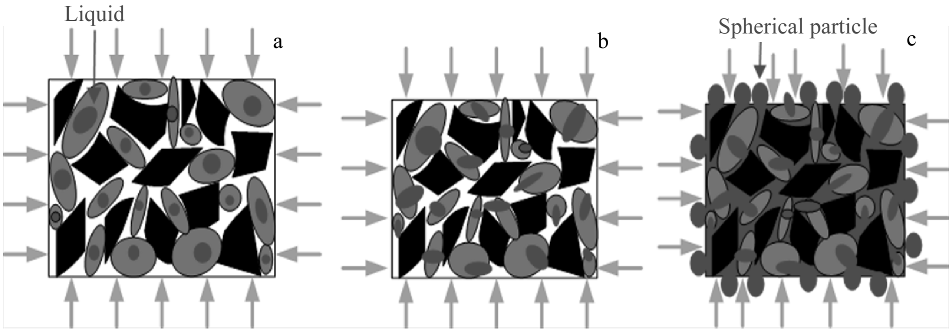

Fig.6 Proposed densification mechanism of semisolid HIP: (a) liquid phase appears, (b) liquid phase overflows, and (c) spherical particles appear

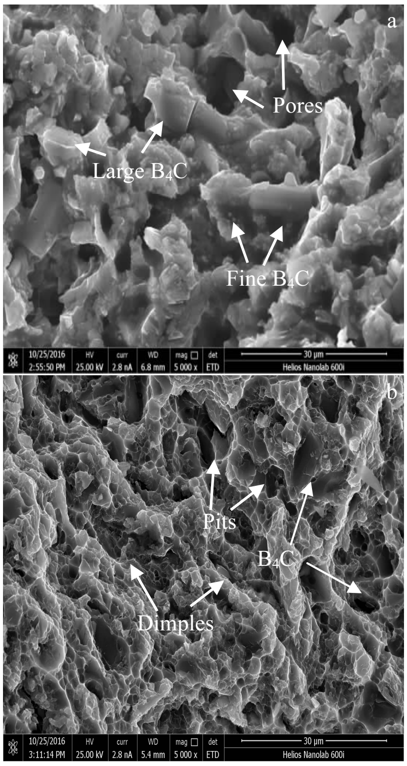

Fig.7 Fracture surface of Al/B_4C composites prepared by different processes: (a) vacuum sintering and (b) semisolid HIP

high plasticity, the presence of pits often indicates deterioration in plasticity. The mechanical properties of Al/B_4C composites depend on the dimples and pits. When the pits are widely distributed in the fractures, the plastic property of composites will be deteriorated. In this study, the plasticity is reduced because of the high content of B_4C, but the tensile strength is greatly enhanced by the addition of mico-B_4C particles.

3 Conclusions

1) The combination between B_4C particles and the Al matrix can be effectively enhanced by the semisolid HIP process. Although the tensile strength can reach up to 300 MPa, the ductility of the semisolid HIP Al/B_4C composite is poor.

2) The B_4C particles can be barely observed on the surface of Al/B_4C composite. The main reason is that the Al liquid forms during the semisolid HIP process and then covers the outer surface of composites.

3) The density of Al/B_4C composites is greatly improved by semisolid HIP process. The densification mechanism can be described by three steps and is mainly related to the aluminum liquid flowing into the internal gaps under the semisolid HIP condition.

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半固态热等静压 Al/B_4C 复合材料液相流动行为与致密化机理

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摘　要：为了制备具有高密度的铝基碳化硼材料，采用粉末冶金半固态热等静压方法制备了质量分数为30%碳化硼的铝基碳化硼复合材料，采用 WANCE100 型材料力学性能试验机和 SIRION200 型扫描电镜研究了复合材料的力学性能及显微形貌。结果表明：半固态热等静压工艺可制备接近理论密度的 Al/B_4C 复合材料；虽然 Al/B_4C 材料抗拉强度可提升至约 300 MPa，但过高碳化硼含量也使得该材料脆性特征十分明显；同时采用间接的方法观察到了半固态工艺过程中生成的液相，该液相不仅可改善碳化硼颗粒与铝基体的结合，在高温高压下液相的流动还起到填充复合材料内部空隙的作用。半固态热等静压工艺过程中产生的液相是复合材料密度和力学性能提升的主要原因。

关键词：铝基碳化硼；半固态；液相；致密化机理

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