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Fabrication and characterization of $SiC_w/MoSi_2$ composite from COSHSed powder

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Abstract SiC whisker reinforced $MoSi_2$ composite has been successfully fabricated by hot-press sintering from ''chemical oven'' combustion synthesized powder. After sintering, a uniform dispersion of SiC whiskers is obtained in the $MoSi_2$ matrix. The Vickers hardness, flexural strength and fracture toughness of the $SiC_w/MoSi_2$ composite are 11.15 GPa, 457 MPa and 6.20 MPa·m$^{1/2}$, increased by 26.1%, 134.3% and 47.3% as compared to $MoSi_2$ matrix, respectively. At last, the mechanism of mechanical properties improvement was also proposed.

Introduction

$MoSi_2$ has attracted great research interest due to its rather low density (6.28 g/cm^3), high melting point, high electrical conductivity and very good oxidation resistance at high temperature, even in very aggressive environments [1–4]. It is useful in such applications as high temperature heating elements and possible structural parts at elevated temperature. However, monolithic $MoSi_2$ exhibits extreme brittleness and poor impact strength at lower temperature, and has low strength and creep resistance at elevated temperature (>1200 °C). Thus, it is essential to increase the room temperature fracture toughness, high temperature strength and creep resistance. Some significant improvements have been obtained through the addition of SiC whiskers or particles to $MoSi_2$ matrix [3, 5–20]. And the mechanical properties of SiC whiskers reinforced $MoSi_2$ composite are better than SiC particles reinforced $MoSi_2$ composite [3, 7–9]. Therefore, SiC whisker-reinforced $MoSi_2$ composites are considered as excellent candidates for high temperature applications.

Although numerous attempts to introduce artificial SiC phase into $MoSi_2$ have been made, only a limited number of studies have addressed their introduction via in situ reactions, and these studies only focus on the introduction of SiC particles [10–17]. These reactions offer the possibilities of generating stable second-phase dispersions in addition to eliminating undesirable interface between second phase and matrix during processing. The interface between the particles and matrix is often a potential source of weakness owing to the very different thermal expansion coefficient.

Combustion synthesis, or Self-propagating high temperature synthesis (SHS) is a technique for producing ceramics, intermetallic, and composite materials. The advantage in the SHS process lies in its low cost, and, with high temperature deformation as a consideration, in the ability to form powders with very fine second phase particles or whiskers. At present, only a limited number of studies reported synthesis of $SiC/MoSi_2$ composite powders by SHS process, and unluckily, these studies only focus on SiC particles reinforced $MoSi_2$ composite powders [18–20]. There are two reasons. Firstly, it is difficult for SiC whiskers to evenly distribute in the reactant mixtures and the form introduction of SiC whiskers in $MoSi_2$ matrix is a problem. Secondly, for the case of some systems, such as $MoSi_2$-SiC, ignition is impossible without addition activation. Primarily this is due to the thermodynamic limitation, i.e. a low

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reaction enthalpy or the relatively low adiabatic combustion temperature of these systems.

Recently, a novel process, referred to in the literature as chemical oven SHS (COSHS) [21] was developed to prepare some powders that cannot be obtained through conventional SHS. In the COSHS mode of combustion, the reactant pellet outside is ignited at upper surface by a high-energy heat input and layerwise combustion occurs at a definite rate of wave propagation. The reactant pellet inside is warmed up by the heat given out from the reaction outside and then ignited at the bottom surface contacted with the reactant outside. Because of the increased reaction temperature and long-time at high temperature, the reaction is carried out thoroughly and impurities are evaporated completely. In this work, SiC whiskers reinforced $MoSi_2$ composite powders were produced from the mixing powders of Mo, Si, carbon black and Si_3N_4 whiskers via COSHS process. The bulk of this paper will discuss the mechanical properties of composite made from COSHSed Powder, consolidated by hot pressing.

Experimental

A 98.5% of pure Mo powder with a particle size range 2–5 μm, 99.4% of pure Si powder with an average size of 10 μm and 99.9% of pure carbon black with an average size of 6 μm. are used as starting raw materials in this study. The Si_3N_4 whiskers were obtained from our research group by SHS process [22]. The samples were prepared according to the composition of 15vol% SiC. The mixtures were combusted via COSHS process in a steel chamber under 0.5 MPa Ar (99.9 wt%) pressure. For sintering studies, the as synthesized fragile mass was milled for 10 minutes in a ball mill. The details of the COSHS technique for the in-situ $SiC_w/MoSi_2$ powder preparation have been published elsewhere [23]. The milled powder was compressed into the form of cylindrical pellets of 30 mm diameter and 10 mm in height by applying a load of 5 MPa. The green pellets were hot pressed at 1700 °C under a pressure of 25 MPa in Ar atmosphere.

The morphologies of sintered products were studied by using scanning electron microscopy (SEM) with an energy dispersive X-ray spectrometer (EDS). Sintered sample densities were measured by the Archimedes method. The Vickers hardness (H_V) and fracture toughness (K_{IC}) were measured on polished specimens using Vicker's diamond indentor under 98N for 15 s. K_{IC} values were calculated by using the equation reported by Anstis et al. [24]. The flexural strength was measured at room temperature using the three-point bending test with a span length of 20 mm and a cross-head speed of 0.5 mm/min.

Results and discussion

Microstructure

The XRD result shows that the as-prepared powder is mainly composed of $MoSi_2$ and SiC phases. Besides the mainly two phases, trace $Mo_{4.8}Si_3C_{0.6}$ (Nowotny phase) also can be observed. SEM photo and EDS result show that SiC whisker is formed during this COSHS process. The results of as-prepared powder by COSHS technique have been published elsewhere [23].

SEM micrographs of polished surface and fractured surface of $SiC_w/MoSi_2$ are shown in Figs. 1 and 2, respectively. In Fig. 1, besides the bright matrix phase, there is a black rodlike phase. Semi-quantitative analysis (EDS) shows that it consists of C (36.7 at%), Si (52.8 at%) and Mo (10.5 at%). This could probably be SiC whisker. From Fig. 1, it can be observed that SiC whiskers are homogeneously distributed in $MoSi_2$ matrix from both the polished surface and fractured surface.

Mechanical properties

The mechanical properties of fabricated composites are given in Table 1. For comparison, the mechanical properties of monolithic $MoSi_2$ are also listed in this table. Significant improvement in hardness has been obtained by incorporation SiC whisker into $MoSi_2$. This may be due to the high hardness of SiC over that of $MoSi_2$. Table 1 also shows the flexural strength and fracture toughness results at room temperature of $SiC_w/MoSi_2$ composite and monolithic $MoSi_2$. The flexural strength and fracture toughness results of $SiC_w/MoSi_2$ composite are 457 MPa and 6.2 MPa·m$^{1/2}$, increased by approximately 134.3% and

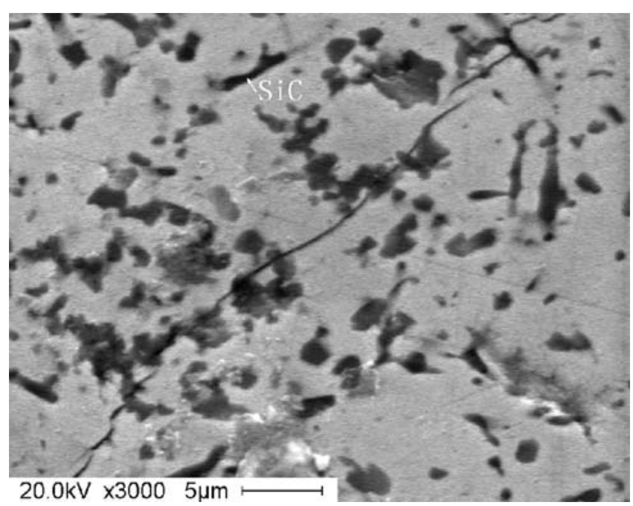

Fig. 1 SEM micrograph of the propagation of crack in the polished surface of $SiC_w/MoSi_2$ composite

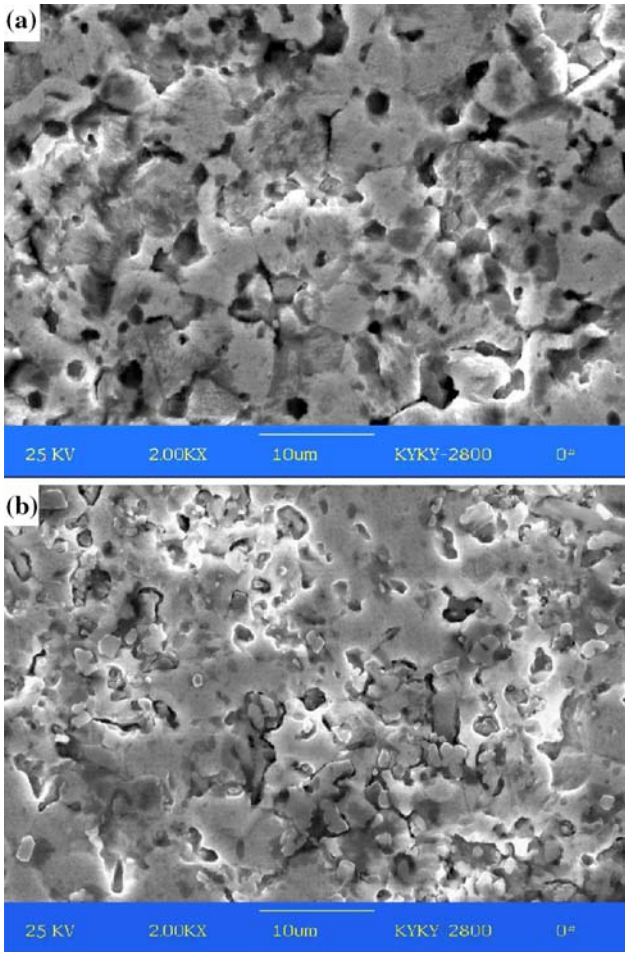

Fig. 2 SEM micrograph of the etched surface of (**a**) $SiC_w/MoSi_2$ composite and (**b**) monolithic $MoSi_2$

Table 1 Mechanical properties of sintered products

Materials	Relative density (%)	Microhardness (GPa)	Flexural strength (MPa)	Fracture toughness ($Mpa \cdot m^{1/2}$)
$MoSi_2$	96.5	8.84	195	4.21
$SiC_w/MoSi_2$	95.6	11.15	457	6.20
20%$SiC_w/MoSi_2$[9]	95.2	13.94	389	5.5

47.3% as compared to $MoSi_2$ matrix, respectively. These results are also better than the results of 20%vol $SiC_w/MoSi_2$ [9] by artificial process, because undesirable interface between second phase and matrix can be eliminated via in situ process. It can be concluded that the addition of SiC whiskers via in situ process significantly improved the flexural strength and fracture resistance of the $MoSi_2$.

Figures 2 and 3 show the etched and fractured surface of $SiC_w/MoSi_2$ composite and monolithic $MoSi_2$, respectively. The average $MoSi_2$ grain sizes of $SiC_w/MoSi_2$ and monolithic $MoSi_2$ were about 3.3 μm and 7.2 μm. It can be clearly observed from Figs. 2 and 3 that $MoSi_2$ grain size of $SiC_w/MoSi_2$ is smaller than monolithic $MoSi_2$, which is benefit to the improvement of flexural strength and fracture toughness.

Figure 1 shows the propagation of crack in the polished surface of $SiC_w/MoSi_2$ composite. With the existence of SiC whiskers in $MoSi_2$ matrix, clear deflection of crack by tilting and twisting around SiC whiskers is observed. This resulted in fracture surfaces that contained holes and mounds in regions in which deflection occurred continuously by tilting around the SiC whiskers (Fig. 3b). The deflection of crack would lead to the increase of fracture toughness. And it is also observed that the whiskers bridge the crack in Fig. 1. Furthermore, holes and outcrops of whiskers by pullout can be seen from the fractured surface in Fig. 3b. Consequently, the traction force of bridging whiskers could efficiently stop the propagation of crack and enhance the fracture toughness.

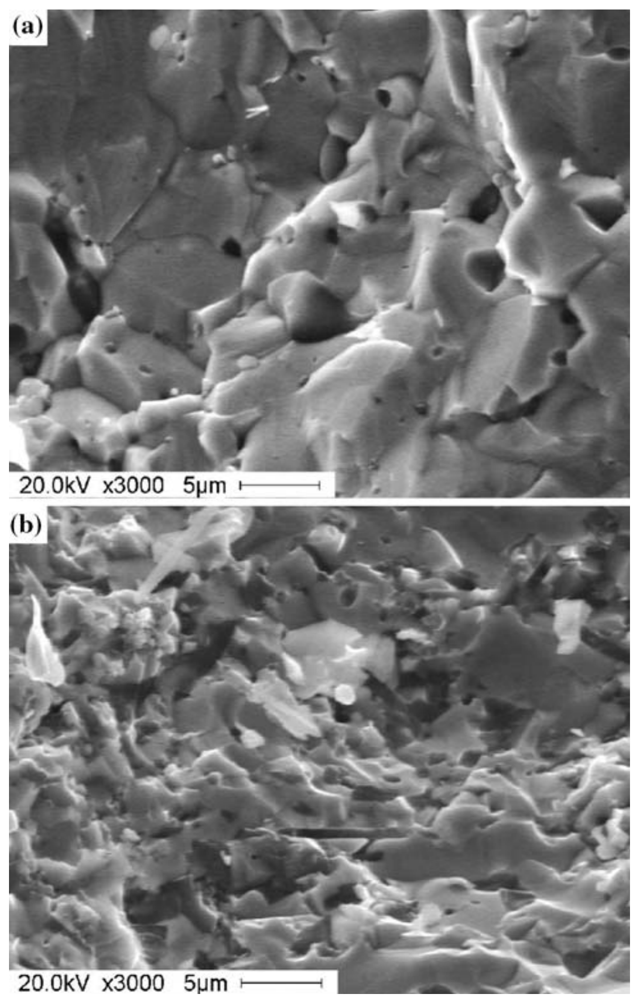

Fig. 3 SEM micrograph of the fractured surface of (**a**) $SiC_w/MoSi_2$ composite and (**b**) monolithic $MoSi_2$

Conclusions

SiC whisker reinforced $MoSi_2$ composite has been successfully fabricated by hot-press sintering from ''chemical oven'' combustion synthesized powder. Increased microhardness, flexural strength and fracture toughness are achieved by the introduction SiC whiskers into $MoSi_2$ matrix. Particle size decrease, cracks deflection and whiskers pullout and bridging are probable mechanism responsible for this behavior.

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