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In situ fabrication and properties of 0.4MoB-0.1SiC-xMoSi$_2$ composites by self-propagating synthesis and hot-press sintering

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ABSTRACT

Mo, Si and B_4C powders were used to fabricate 0.4MoB-0.1SiC-xMoSi$_2$ composites by self-propagating high-temperature synthesis (SHS) and hot pressing (HP). The effects of $MoSi_2$ content (x = 1, 0.75, 0.5 and 0.25) on phase composition, microstructure and properties of the composites were investigated. The results showed that the 0.4MoB-0.1SiC-xMoSi$_2$ composite exhibited Vickers hardness of 10.7–15.2 GPa, bending strength of 337–827 MPa and fracture toughness of 4.9–7.0 MPa m$^{1/2}$. The fracture toughness increased with the increasing volume fraction of MoB and SiC particles which were promoted by the toughening mechanisms, such as crack bridging, cracks deflection and crack branching. Moreover, the electrical resistivity showed an increasing trend with decreasing volume fraction of $MoSi_2$.

1. Introduction

Molybdenum disilicide ($MoSi_2$) as structural and functional material has received applications in high temperature environment [1–4] due to its good oxidation resistance (> 1600 °C), high melting point (2030 °C) and low density (6.24 g/cm^3). However, its fracture toughness at room temperature and creep resistance at high temperature are not satisfactory [5–8]. The addition of alloying elements (such as Nb, Al, Ta, and Al) or their synthetic products have been regarded as the promising routes to improve the toughness of structural silicides. For instance, the fracture toughness of, Al-alloyed $MoSi_2$ matrix composites was 3.7 MPa m$^{1/2}$ [9], $NbSi_2$/$MoSi_2$ duplex crystals was 4.1 MPa m$^{1/2}$ [10], Nb and Al co-substituted $MoSi_2$ was 4.0 MPa m$^{1/2}$ [11].

Numerous methods of second phase strengthening have been recently developed to toughen and strengthen $MoSi_2$, such as addition of continuous ceramic fibers, discontinuous whisker, second particles and ductile phases [12–14]. Reinforcements of SiC [15,16], ZrB_2 [16], Si_3N_4 [17,18], BN [18], Al_2O_3 [19] can increase fracture toughness by introducing extrinsic toughening mechanisms such as fiber strengthening, crack deflection and branching [20]. Wang et al. [21] investigated the $MoSi_2$-SiC_w composites and reported the maximum fracture toughness and flexural strength were 5.8 MPa m$^{1/2}$ and 646 MPa respectively. Magnanin et al. [22] fabricated the SiC-$MoSi_2$ composites and the reported fracture toughness and bending strength were 6.80 MPa m$^{1/2}$ and 242 MPa. Thus, the addition of SiC particles to $MoSi_2$ matrix can be considered as an effective and important route to enhance the mechanical properties of $MoSi_2$ matrix composites.

Mo-B phases were employed as structural materials and showed good electrical conductivity, which made these phases promising candidates to reinforce $MoSi_2$ materials [23]. Costa e Silva et al. [24,25] synthesized MoB-$MoSi_2$ and Mo_2B_5-$MoSi_2$ composites using in situ displacement reactions with Mo_5Si_3 and boron particles by hot-pressing (HP) at 1400 °C and $MoSi_2$-boride composites displayed significantly high hardness at room temperature and slight improvements in fracture toughness than pure $MoSi_2$. Moreover, the grain size of the silicide and boride were substantially finer in MoB-$MoSi_2$ than Mo_2B_5-$MoSi_2$ composites. Schneibel et al. [26] studied the mechanical properties of $MoSi_2$-MoB and reported the flexure strength and fracture toughness of 320 MPa and 3 MPa m$^{1/2}$, respectively. Nevertheless, there are opportunities to incorporate multiphase reinforcements to design $MoSi_2$-based composites with superior fracture toughness.

In situ multiphase MoB-SiC-$MoSi_2$ composites offers an interesting alternative to $MoSi_2$-based materials. The objective of this study is to research the in situ fabrication of multiphase 0.4MoB-0.1SiC-xMoSi$_2$ (x = 1, 0.75, 0.5, and 0.25) composites using self-propagating high-temperature synthesis (SHS) and hot-pressing. The influences of starting composition on the microstructure and mechanical properties of the MoB-SiC-$MoSi_2$ composites have been reported.

2. Experimental

Molybdenum (−300 mesh, 99.9% purity, Zhuzhou cemented

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carbide group Co. Ltd, China), silicon (3.0–5.0 μm, 99.9% purity, WODETAI (Beijing) Co. Ltd) and carbide tetraboride (1.0–10.0 μm, 99.9% purity, Aladdin industrial, Co. China) powder mixtures were prepared with four different nominal compositions of 0.4MoB-0.1SiC-1$MoSi_2$ (MBS1); 0.4MoB-0.1SiC-0.75$MoSi_2$(MBS0.75); 0.4MoB-0.1SiC-0.5$MoSi_2$ (MBS0.5); 0.4MoB-0.1SiC-0.25$MoSi_2$ (MBS0.25). The raw materials were mixed in a planetary ball mill for 120 min at 450 rounds per minute using agate jar and balls, and the ball to powder ratio (BPR) is 2:1, and alcohol was chosen as the milling medium. Then the milled mixtures were dried and cold-pressed into cylindrical compacts of 16 mm in diameter and 15 mm in height under an applied pressure of 200 MPa. The combustion synthesis experiments were conducted in a steel combustion chamber in a pure argon (99.99%, 0.1 MPa) atmosphere and experienced a preheating by an energized molybdenum wire with 15 V and 15 A in 1, 2, 4, 5 min, respectively. The method of ignition was heating a molybdenum wire through 25 V and 28 A. The synthesized samples were broken and crush into powders (−200 mesh), which were ball milled for 3 min in impact grinder, then the powder was ball milled for 240 min at 450 rounds per minutes again.

The powders were sintered by hot-pressing at 1500 °C with 10 °C/min and 27.5 MPa for 1.5 h in vacuum (< 6.0 × 10^{-3} Pa). Gac el at. [27] studied feasibility of a composite of SiC whiskers in an $MoSi_2$ matrix and reported the densifying composites was 95.2–97.5% by hot-pressing of 1625 °C and 41.4 MPa. Mitra et al. [28] researched processing-microstructure-property relationships in reaction hot-pressed $MoSi_2$ and $MoSi_2/SiC_p$ composites at 1500 °C for 1 h using a pressure of 26 MPa, and the range of density was 95–98%. Moreover, the planetary ball milling and sintering technology have been studied in the previous works, and the preparation technologies are suitable for densification of $MoSi_2$-maxtix composites (density, > 95%) [29]. The sintered plates (50 mm × 4 mm) were cut into test pieces that measured 10 sheets about 22 mm × 3 mm × 3.5 mm and 1 sheet about 14 mm × 14 mm × 3 mm by wire-electrode cutting.

The combustion temperature was test by infrared temperature apparatuses (Reytek, marathon series made in USA), the sample surfaces were ground and polished to a mirror finish. Surfaces were analyzed by scanning electron microscope (SEM, FEI Quanta TM 250) equipped with energy-dispersive spectroscopy (EDS, Quantax 400-10). The phase compositions were identified by Bruker D8 Advance X-ray diffraction (XRD, Cu Ka (λ = 0.15406 nm)), electrical resistivity(ER) is tested by DC Low-ohm Meter (TH2513A, Changzhou Tonghui Electronic Co, Ltd), the universal-testing was used for bending strength. The volume fraction of each phases was calculated by image pro software.

3. Results and discussion

3.1. Combustion synthesis

The conversion of the Mo, Si, and B_4C powders into 0.4MoB-0.1SiC-$x$$MoSi_2$ composites could be described according to the following reaction during self-propagating high temperature synthesis (SHS):

$$(0.4 + x)\,Mo + (0.1 + 2x)\,Si + 0.1B_4C \rightarrow 0.4MoB + 0.1SiC + xMoSi_2 \tag{1}$$

In Fig. 1 the combustion temperature-time curves of 0.4MoB-0.1SiC-$x$$MoSi_2$ (x=1 (MBS1), 0.75 (MBS0.75), 0.5 (MBS 0.5), 0.25 (MBS0.25)) composites show that the combustion temperature of MBS1 is 1937 K and it increases to 1966 K for MBS0.25, while the variation of combustion temperature is contrary to the theoretical adiabatic temperature (T_{ad}), which is 1921–1895 K from MBS1 to MBS0.25, respectively, before the preheating procedure. The lowest theoretical T_{ad} is only 95 K higher than the critical T_{ad} of 1800 K at the room temperature condition [30,31] and the actual T_{ad} is influenced by compact density, atmosphere and other experimental conditions [23]. Thus, all the green compacts were preheated to ensure the self-prorogating reaction in this study. During SHS reaction, the minimum combustion temperature (1937 K for MBS0.25) exceeded melting point of Si (1685 K), however, the maximum combustion temperature (1966 K for MBS1.0) was lower than the melting point of other components (2890 K for Mo and 2623 K for B_4C). It suggests that the mechanism of combustion synthesis can be characterized with Mo and B_4C dissolving in liquid Si and resulting in precipitation of $MoSi_2$, MoB and SiC particles, namely, the dissolution and precipitation mechanism [32].

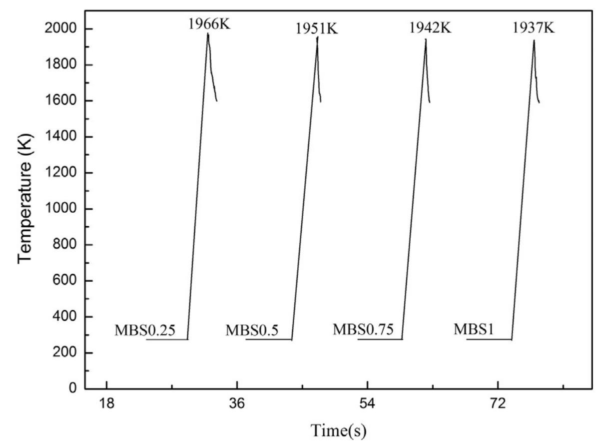

Fig. 1. The combustion temperature-time curve of MoB-SiC-$MoSi_2$ composites with different $MoSi_2$ molar ratio.

X-ray diffraction (XRD) patterns of the SHS-HP products are shown in Fig. 2. The MoB, SiC (moissanite-15R) and $MoSi_2$ peaks are detected. The relative intensities of MoB diffraction peaks increase with decreasing the $MoSi_2$ content from MBS1 to MBS0.25. The strongest peak is from $MoSi_2$ phase (at 2θ = 45.026°) from MBS1 to MBS0.75, while the strongest peak of MBS0.25 is MoB phase (at 2θ = 42.692°). The XRD data confirms that MoB-SiC-$MoSi_2$ composites could be fabricated by this simple and rapid combustion synthesis method (See Supplementary information, combustion synthesis video).

3.2. Microstructure

The BSED images and EDS mapping of MBS0.25 are shown in Fig. 3. The chemical elements of B, C, Mo, Si and O were detected. The I part was marked in Fig. 3a, c, d and e which were in the same position. The I parts showed that the concentration of Si elements was highest, the C elements were densely distributed and the concentration of Mo element was in a low level. On II areas, the concentration of Mo and B elements was higher than on I and III parts, while Si element was lowest. The concentration of Mo and Si was relatively moderate on III part. The

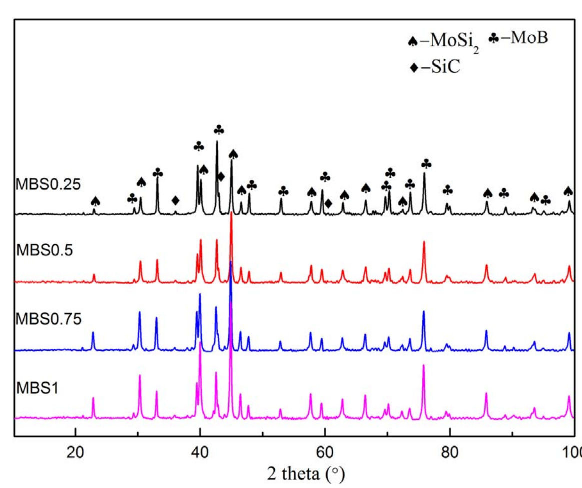

Fig. 2. X-ray diffraction patterns of the combustion synthesis and hot pressing products.