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Synthesis of $SiC_w/MoSi_2$ powder by the ''chemical oven'' self-propagating combustion method

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Abstract

SiC whisker reinforced $MoSi_2$ composite powder has been successfully synthesized by ''chemical oven'' combustion synthesis method. The mixtures of Si and Ti were ignited as chemical oven. XRD result shows that the combustion product is mainly composed of $MoSi_2$ and SiC phases. SEM photo and EDS result show that SiC whisker is formed during this process.

Keywords: A. Powders: solid state reaction; B. Composites; B. Whiskers; D. Silicides; Chemical oven

1. Introduction

$MoSi_2$ (molybdenum disilicide) 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 [3,5,6]. And the mechanical properties of SiC whiskers reinforced $MoSi_2$ composite are better than SiC particles reinforced $MoSi_2$ composite [3]. Therefore, SiC whisker-reinforced $MoSi_2$ composites are considered as excellent candidates for high temperature applications.

Up to now, reinforcements have been introduced primarily by mechanical mixing or in situ synthesis. Disadvantages in the processing techniques lie in the unequal distribution of the reinforcing phase and, more importantly, in the size limits of the reinforcing phase. So preparing of $SiC/MoSi_2$ composite powders can be an important stage for making $SiC/MoSi_2$ composite.

Combustion synthesis, or self-propagating high temperature synthesis (SHS) is a technique for producing ceramics, intermetallic, and composite materials. It utilizes the heat generated by an exothermic reaction to sustain itself in the form of a combustion wave after external ignition. 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 self-propagating high temperature synthesis or mechanical alloying (MA) process, and these studies only focus on SiC particles reinforced $MoSi_2$ composite powders [7–10]. There are two reasons. Firstly, it is difficult for to allow for an even distribution of SiC whiskers in the reactant mixtures and the introduction of SiC whiskers in $MoSi_2$ matrix is a problem. Secondly, for the systems such as $MoSi_2$-SiC ignition is impossible without addition activation. Primarily

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this is due to the thermodynamic limitation, i.e. a low 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) [11,12] 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. SiC whiskers can be introduced by reacting Si_3N_4 whisker with carbon [13,14].

2. Experimental procedures

98.5% pure Mo powder with a particle size range 2–5 μm, 99.4% pure Si powder with an average size of 10 μm and 99.9% pure carbon black with an average size of 6 μm are used as starting raw materials in this study. Also, the mean diameter and length of Si_3N_4 whiskers used were 1–2 μm and aspect ratio of 10–20. The Si_3N_4 whiskers were obtained from our research group by Self-propagating High-temperature Synthesis process [15]. The microstructure of

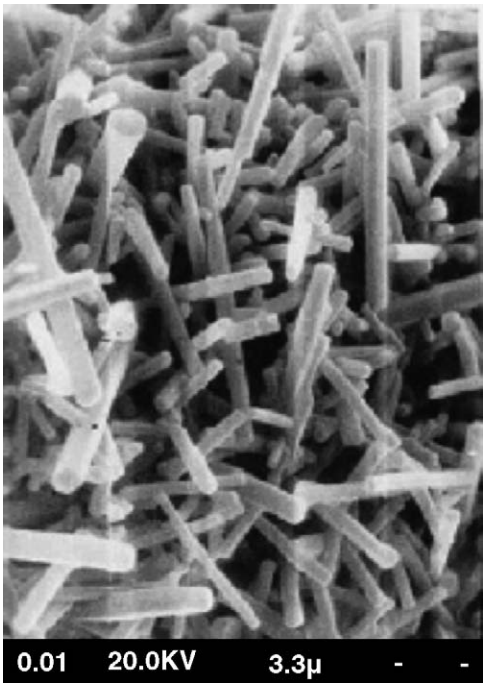

Fig. 1. SEM micrograph of Si_3N_4 whiskers used in this study.

Table 1
Composition of starting powder (argon pressure: 0.5 MPa)

Sample	Method	Phase composition of starting powders (wt.%)			
		Si	Mo	C	Si_3N_4
A	SHS	36.93	63.07	0	0
B	SHS	35.54	60.68	0.78	3
C	COSHS	32.62	55.71	2.40	9.27

Si_3N_4 whiskers is shown in Fig. 1. The samples were prepared according to the composition of 0, 5, 15 vol% SiC shown in Table 1. The mixtures were milled with Si_3N_4 balls in absolute ethanol for 24 h. After milling, the resultant slurry was dried and then passed through 60 mesh sieve to obtain an agglomerate-free powder mixture for following two separate SHS procedures.

2.1. Normal SHS procedure

The mixed powders were pressed into a cylindrical graphitic crucible with typical dimensions of 40 mm diameter and 45 mm high. A tungsten coil was placed on the upper surface of the powders. The powders were combusted on a steel chamber under 0.5 MPa Ar pressure. The purity of the Ar gas was reported as 99.9%.

2.2. Chemical oven SHS procedure (COSHS)

The mixtures were put into inner part of the graphite crucible and mixtures of Si and Ti (atomic ratio Si/Ti = 3/5) used as chemical oven were put into outer part shown in Fig. 2. There was carbon felt between inner part and outer part. A tungsten coil located on the upper surface of the powders in outer part. The two mixtures get in touch with each other on the bottom of the graphite crucible. The powders were combusted in a steel chamber under 0.5 MPa Ar (99.9 wt.%) pressure.

The combustion wave was determined from the output of a w-5%Re/w-26%Re thermocouple placed inside a small hole drilled in the side of each specimen. The samples were

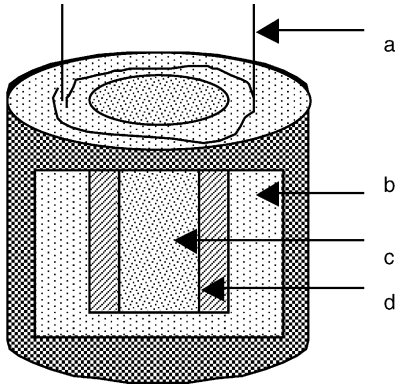

Fig. 2. Schematic of experimental apparatus used for chemical oven combustion synthesis (a) W coil; (b) 3Si + 5Ti (atomic ratio); (c) 2Si + Mo (atomic ratio); (d) carbon felt.

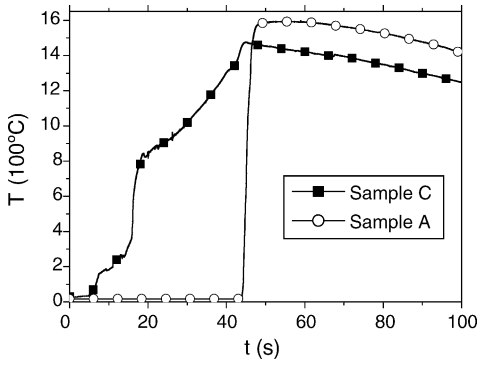

Fig. 3. Temperature profiles of samples A and C.

characterized by powder X-ray diffraction (XRD), employing a scanning rate of 0.05° s^{-1} with a 2θ range from 10° to 70°, using a Rigaku Dmax γA X-ray diffractometer with graphite monochromatized Cu Kα radiation (λ = 1.54178 Å). The morphologies of combustion products were studied by using scanning electron microscopy (SEM).

3. Results and discussion

The temperature profiles obtained both in SHS and COSHS are indicated in Fig. 3. Except Sample B, the other two samples all can be reacted under conditions shown in Table 1. Though sample B has contain less carbon black and Si_3N_4 whisker than sample C, it cannot carry out in normal SHS, because the heat given out by the reaction between Si and Mo is not enough for both sustaining propagation and completing the reaction between carbon black and Si_3N_4 whisker. In COSHS procedure, additional heat given by the reaction between Si and Ti make the reaction between carbon black and Si_3N_4 whisker completely finished. Compared to the single heating process of sample A, it can be clearly seen that the heating process of sample C is divided to three steps from Fig. 3. Firstly, the reactant had been heated up to ~370 °C by the ''chemical oven'', then the reactant had been ignited and temperature grew up at dramatically rate, at last when the temperature touched ~840 °C the heating rate is lowered because some

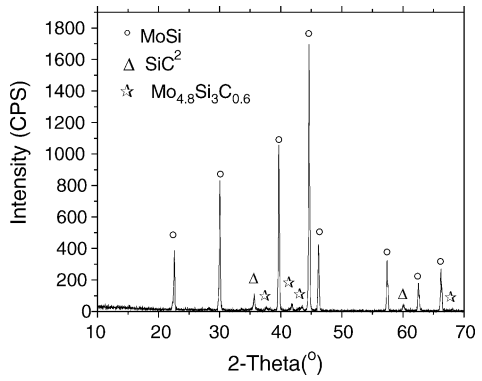

Fig. 4. XRD pattern of sample C.

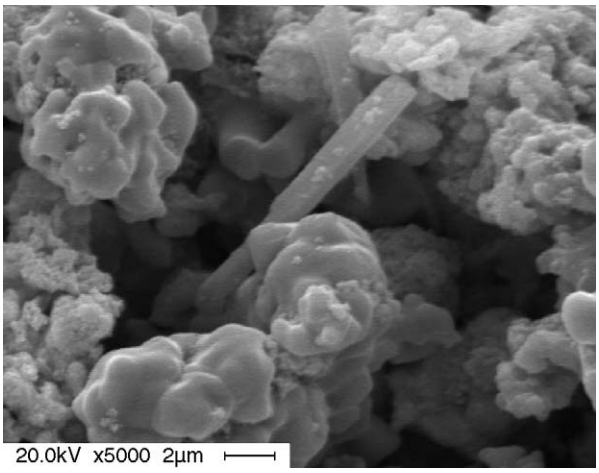

Fig. 5. SEM micrograph of sample C.

endothermic processes had been activated. The maximum temperatures of sample C recorded in COSHS procedure is 1477 °C while the maximum temperature of sample A recorded in normal SHS is 1598 °C, higher by 121 °C.

XRD pattern of Sample C is shown in Fig. 4. It shows that there are no residue Si and C phase in sample C, so the reaction is completely finished. From Fig. 4, it can be seen $MoSi_2$ is the major phase and SiC is the second phase of the composite. Besides the mainly two phases, trace $Mo_{4.8}Si_3C_{0.6}$ (Nowotny phase) also can be observed. And there is a great possibility for $Mo_{4.8}Si_3C_{0.6}$ to be reinforcement second phase [16].

Fig. 5 shows SEM photograph of sample C. $MoSi_2$ grains, which agglomerated and formed a three dimensional network structure, were observed in this sample. Besides the main particle phase, there is a rod-like phase in Fig. 5. Semi-quantitative analysis (EDS) shows that it consists of C (36.1 at%), Si (45.3 at%), Mo (8.2 at%) and N (10.4 at%). This could probably be SiC whisker with Si_3N_4 and $Mo_{4.8}Si_3C_{0.6}$.

4. Conclusion

SiC whisker reinforced $MoSi_2$ composite powder has been successfully synthesized by the ''chemical oven'' combustion synthesis method. SiC whisker is obtained from Si_3N_4 whisker. The mixed powders of Si and Ti were used as chemical oven to enhance SHS procedure for making the process available. It can be concluded that this process of combustion synthesis is a potential useful method for preparing some powders that cannot be obtained through conventional SHS.

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