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An investigation on the in situ synthesis–sintering and mechanical properties of $MoSi_2$–xSiC composites prepared by spark plasma sintering

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ABSTRACT

Composites of $MoSi_2$–x wt.% SiC (x = 5, 10, 15, 20) prepared using spark plasma sintering. The effect of temperature on the in-situ synthesis–sintering was investigated between 1100 °C and 1500 °C. X-ray diffraction patterns showed that at 1100 °C the reactions were incomplete and elementary diffraction peaks of Mo, Si and C still exist. With an increase in temperature from 1100 to 1300 °C the reactions were performed completely. The study showed that the sintering ability at higher temperature at the presence of enough mechanical pressure was better because the heat released from the reaction between Mo, Si and C causes higher temperature than the melting point of Si (1410 °C). Consequently the silicon would melt during the heating process. The molten Si can strengthen the interconnections and it has higher diffusion rate. Therefore, due to the liquid phase sintering and at the presence of mechanical pressure, the sintering ability at higher temperature is better than lower temperature. Scanning electron microscopy showed that with the addition of carbon, there was no silica phase in the microstructure of the synthesized samples, due to the formation of SiC. Therefore, it can be noted that the addition of carbon leads to better mechanical properties due to elimination of silica phase.

1. Introduction

The interest in $MoSi_2$ is due to its high melting point, good oxidation and corrosion resistance and low density [1]. However, $MoSi_2$ has its intrinsic limitation in properties such as low ductility at low temperature. Indeed the mechanical behavior of $MoSi_2$ can be divided into three ranges [1]: strong and brittle (up to approximately 1000 °C), strong and ductile (1000–1250 °C) and weak and ductile (above 1250 °C) [2]. It is known that during the processing of $MoSi_2$ using hot pressing, hot isostatic pressing and spark plasma sintering (SPS) or other processes, SiO_2 forms at grain boundary or inside the grains. This SiO_2 phase leads to the deterioration of the mechanical properties at both ambient and elevated temperatures. Although several investigators reported the observation of silica in $MoSi_2$ based materials, but there is no agreement on the morphology and distribution of silica in $MoSi_2$ and the effect of processing variables on these characteristics. A number of researchers claimed that SiO_2 wets the grain boundary or it concentrates in triple junction [3,4], while others observed globular silica particles both at grain boundary and inside the grains [5–7].

The introduction of reinforcement improves mechanical properties of $MoSi_2$. For instance, many reinforcements have been investigated over the past years, these reinforcements include metals (Nb, W, Ti), nitrides (Si_3N_4, AlN), oxides (Al_2O_3, ZrO_2), carbides (TiC, SiC, ZrC) and borides (ZrB_2, TB_2) [3–5]. Attempts have been made to improve mechanical properties of $MoSi_2$ including: the introduction and control of second phase, tailoring interface properties, microstructural control and alloying [1]. The main criteria for the selection of second phase reinforcement have been reviewed in details elsewhere [5–9]. The most important issues are mechanical properties, coefficient of thermal expansion (CTE), density, chemical compatibility and interfacial characteristic. Among these various reinforcements, SiC is considered to be significantly effective due to its good compatibility with $MoSi_2$.

Many processing methods have been used to prepare $MoSi_2$ composite with SiC particle. These methods include melting process [10], plasma spray deposition [11], mechanical alloying, self propagating high temperature synthesis, hot press, hot isostatic press, spark plasma sintering (SPS) of Mo, Si and C powders [12–15] or reaction of $MoSi_2$ powder with C [16,17]. Except SPS method, the limitations of all these methods lie in that they are time consuming and/or the products are porous. In recent decades SPS has been widely employed for fabrication of many ceramics, metals, intermetallic compounds and different composites [18–20]. In this method the raw powders in a carbon die are pressed uniaxially and direct current (DC) is applied simultaneously. At early stage of the process, the powders are heated by spark discharge between particles and the carbon die is also heated by joule effect, so the powders are heated from the inside and outside [18,19]. Therefore this method leads to shortening the sintering time and good densification.

In the present study we used spark plasma sintering (SPS) in order to synthesis and sinter $MoSi_2$–x wt.% SiC (x = 5, 10, 15, 20) composites using Mo, Si and C powders in one step. The effect of temperature, mechanical pressure and composition on the synthesis, densification,

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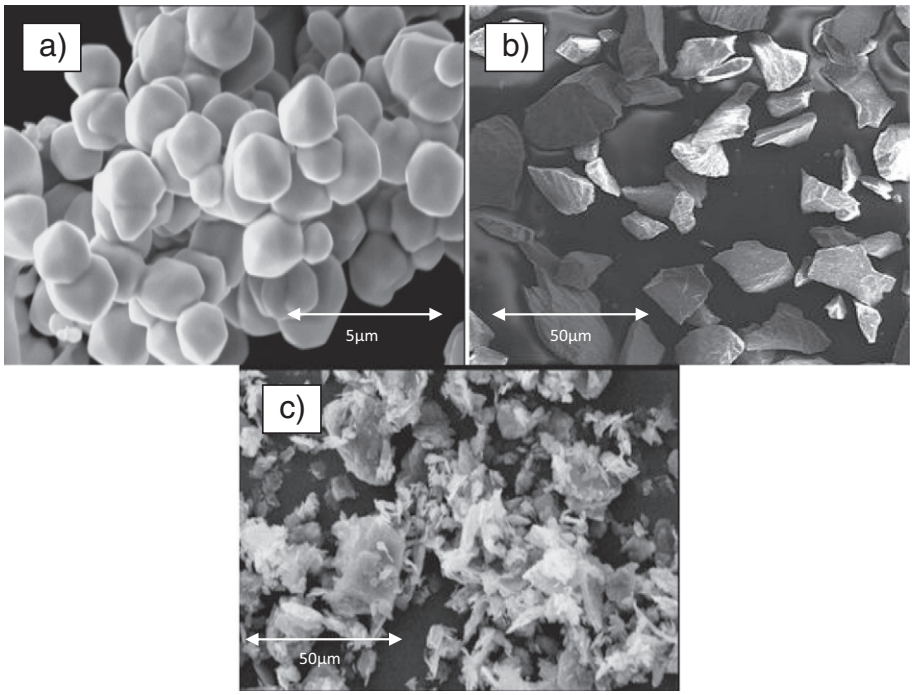

Fig. 1. SEM image of the raw materials used in this study, a) Mo, b) Si and c) C powder.

sintering, microstructure and mechanical properties of samples was also investigated.

2. Experimental

2.1. Mixing and drying

Powders of molybdenum, silicon and graphite were used as starting materials with purity of 99.9%. Fig. 1 shows the scanning electron microscopy (SEM) images of the used raw materials. The atomic ratio of each element is based on the reaction (1):

$$Mo + (x+2)Si + xC = MoSi_2 + xSiC. \quad (1)$$

The powders dispersed in acetones according to stoichiometric ratio of $MoSi_2$–(5, 10, 15, 20) wt.% SiC. The solids loading were approximately 50 wt.% and mixed using a high energy planetary ball mill for 5 h at 200 rpm. The ball to powder ratio was 5:1. After this step, in order to keep constituents from settling, drying was carried out using a hot-plate, while stirring continuously. Then the powders loaded into the graphite die and the graphite die containing raw powders placed inside the SPS vacuum chamber and the powders compacted into a green body using 10 or 15 MPa uniaxial pressure depending on the maximum of pressure.

2.2. Synthesis and sintering of the compacted bodies

The second step of the process, consisting of simultaneously reaction and consolidation of the mixed powder, performed using SPS apparatus. The synthesis–sintering process was performed under high pulsed direct current (between 1000 and 6000 A) in a vacuum atmosphere (8 Pa). Based on previous works by authors [18,19], half of the pressure

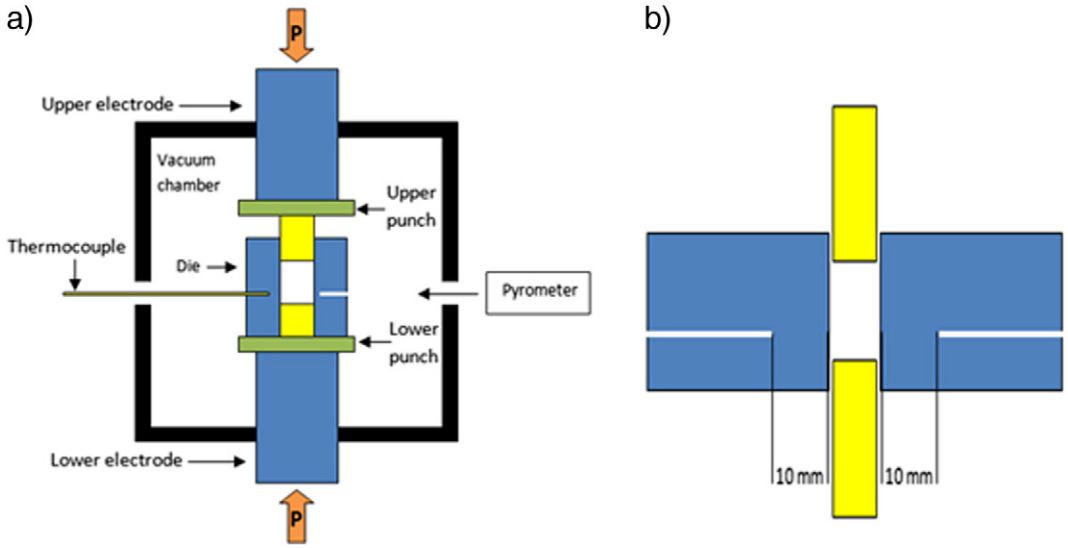

Fig. 2. Schematic of spark plasma sintering apparatus, a) vacuum chamber, b) the position of thermocouple and pyrometer.