Available online at www.sciencedirect.com

SciVerse ScienceDirect

Ceramics International 38 (2012) 6895–6898

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Short communication

In-situ preparation of SiC–$MoSi_2$ composite by microwave reaction sintering

Jianguang Xu*, Fang Chen, Feng Tan

Hunan Provincial key defense laboratory of high temperature wear-resisting materials and preparation technology, Hunan University of Science and Technology, Xiangtan 411201, Hunan, PR China

Received 7 March 2012; received in revised form 20 April 2012; accepted 21 April 2012
Available online 4 May 2012

Abstract

A 20% volume SiC reinforced $MoSi_2$ composite has been successfully in-situ prepared by precisely controlling the microwave reaction sintering process. After sintering, a uniform dispersion of SiC particles was obtained in the $MoSi_2$ matrix from the reaction of Mo, Si and C powders. The sintered product is a composition of $MoSi_2$, $Mo_{4.8}Si_3C_{0.6}$ and SiC. The relative density, flexural strength, Vicker's hardness and fracture toughness of the composite are 91.5%, 251.29 MPa, 7.86 GPa and 8.17 MPa $m^{1/2}$, respectively. We found the heating procedure is critical in the microwave reaction sintering process to obtain dense SiC–$MoSi_2$ composites.

Keywords: A. Sintering; B. Composites; C. Mechanical properties; D. Silicides

1. Introduction

$MoSi_2$ has attracted great research interests due to its rather low density (6.28 g/cm^3), high melting point, high electrical conductivity and excellent oxidation resistance at high temperature [1–4]. However, monolithic $MoSi_2$ shows extreme brittleness and poor impact strength at lower temperature, and has low strength and creep resistance at elevated temperature (> 1200 °C). It has been demonstrated that compounding with a second phase or alloying with other elements is a valid method to strengthen and toughen $MoSi_2$. Some significant improvements have been gained through the addition of SiC to $MoSi_2$ matrix [5–13]. Therefore, SiC-reinforced $MoSi_2$ composites are considered as excellent candidates for the high temperature applications.

Numerous approaches to introduce SiC phase into $MoSi_2$ have been developed recently. Among them, only a few studies have been involved in in-situ reactions [7–14], by which the stable second-phase dispersions could be generated to remove the undesirable interface between the second phase and matrix. This interface often results in the weakness of the composite for the very different thermal expansion coefficient of $MoSi_2$ and SiC. Especially, when the surface of particles is not clean, cracks may propagate from the interface.

Hot pressing is a conventional process to prepare $MoSi_2$ and $MoSi_2$-based materials [15,16], in which powders are sintered at substantially high temperature for a long time. Therefore, it would be very meaningful to develop a fast, clean and energy-efficient technology, to make $MoSi_2$/SiC composite. Recently, we focused our research interests on the microwave irradiation, which directly interacts with the particulates within the pressed compacts and thereby provides rapid volumetric heating. Besides its high efficiency, the uniform heating of macrowave could also minimize the problems such as local microstructural coarsening, as a result yielding better properties.

There are a few examples of microwave reaction sintering have been reported. In 2002, Peelamedu [17] synthesized $NiAl_2O_4$ with good mechanical property by microwave-assistant reaction sintering which provided better sintering rate from the non-isothermal environment. Then Ganesh [18] claimed a nanocrystal $MgAl_2O_4$ has been made by using microwave sintering. In 2003, Panneerselvam [19] attempted to prepare $MoSi_2$–SiC from the element reactants Mo, Si and C by the microwave in-situ

*Corresponding author. Tel.: +86 731 58290847; fax: +86 731 58290043.
E-mail address: jgxu@163.com (J. Xu).

0272-8842/$36.00
http://dx.doi.org/10.1016/j.ceramint.2012.04.072

sintering. Unfortunately, it is difficult to get compacted product. Compared to the normal reaction sintering, microwave reaction sintering is harder to control for its violent reaction condition resulted from the rapid heating, which is often a few times faster than that of normal reaction sintering. Based on this, we think it would be necessary to apply different heating rates at different sintering stages during the microwave reaction sintering to obtain desired products, and this has been proved in our recent research. Here, we report the example to successfully make compacted $SiC/MoSi_2$ composites from the element reactants, by controlling the microwave sintering precisely.

2. Material and methods

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 20 μm are used as starting raw materials in this study. They were mixed according to the composition of 0–20 vol% SiC and 100–80 vol% $MoSi_2$. The mixture was milled for 3 h in a stainless steel vial of volume 125 ml with sialon balls (5 mm diameter) under argon. The weight ratio of the balls to the powders' mixture was 10:1. The rotation speed was 300 rpm.

The as-milled powder was sieved and uniaxial pressed at 200 MPa. The pressed green compact was placed in a MW-L031FHV microwave sintering vacuum oven and sintered at 1400 °C for 20 min. The as-received samples were machined into 3 mm × 4 mm × 36 mm bars. The densities were measured by the Archimedes method. The flexural strength was measured at room temperature using the three-point bending test with a span length of 30 mm and a cross-head speed of 0.5 mm/min. The Vickers hardness (H_V) and fracture toughness (K_{IC}) were measured on polished specimens using Vicker's diamond indenter under 294 N for 15 s. K_{IC} values were calculated by using the equation reported by Anstis [20].

3. Results and discussion

XRD pattern of the sintered sample is shown in Fig. 1. $MoSi_2$ (JCPDS:41–0612) is the major phase and SiC (JCPDS:29–1129) is the second phase of the composite. Besides those two main phases, trace $Mo_{4.8}Si_3C_{0.6}$ (JCPDS:43–1199, Nowotny phase) also could be observed.

Fig. 2 shows the fractured surface of monolithic $MoSi_2$ and $MoSi_2$–SiC composite. The monolithic $MoSi_2$ was obtained by the same microwave sintering process from Mo and Si powders. There are two types of grains in $MoSi_2$–SiC composite in Fig. 2b. The big grains ranging 2–8 μm, which are $MoSi_2$, contact each other and construct the matrix. The small grains ranging 0.2–1 μm, which are SiC, distributed homogeneously in grain boundaries among $MoSi_2$ grains. Compared to pure $MoSi_2$, the size of $MoSi_2$ grains and pores decreased a lot.

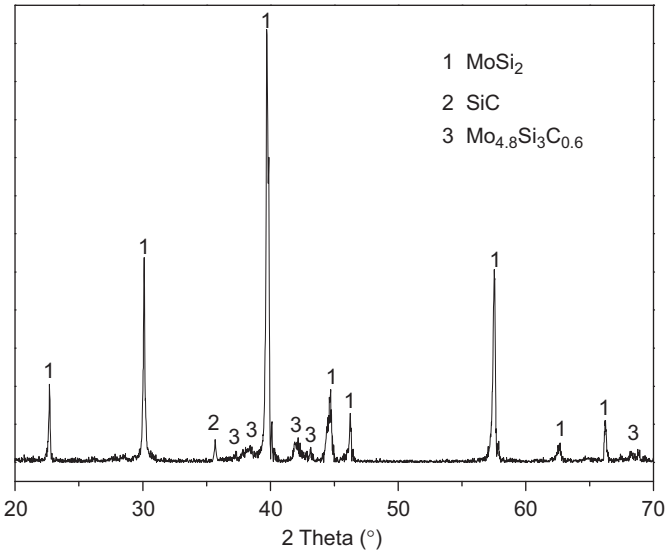

Fig. 1. XRD pattern of the $MoSi_2$–SiC composite.

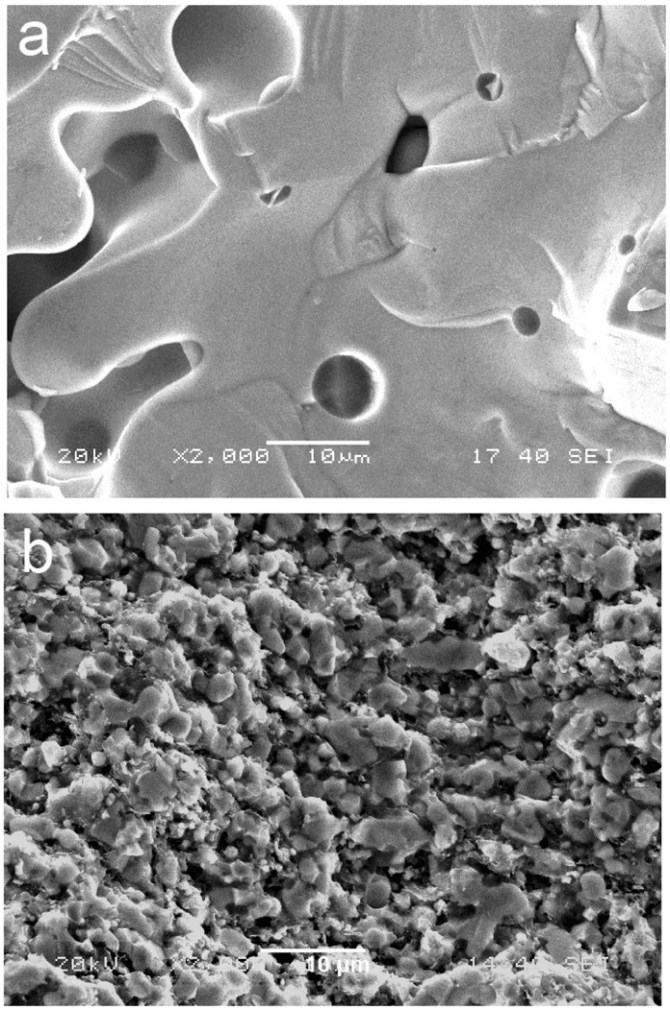

Fig. 2. SEM micrograph of the fractured surface of (a) monolithic $MoSi_2$ and (b) $MoSi_2$–SiC composite.

The densities, mechanical properties of the as-sintered samples are listed in Table 1. The theoretical density of $SiC/MoSi_2$ composites were calculated by the additivity rule of composite materials. The relative density of $MoSi_2$-SiC composite is 91.5%, which is 13.3% more than that of monolithic $MoSi_2$. The element C could help to get rid of SiO_2, which formed on the surface of Si particle. In this way, the diffusion rate of Si during the sintering process would increase and lead to the increase of the density. Furthermore, the flexure strength, the Vicker's hardness and the fracture toughness of the $MoSi_2$–SiC composite are 251.29 MPa, 7.86 GPa and 8.17 MPa $m^{1/2}$, increased by 131.3%, 69.0% and 125.1% compared to monolithic $MoSi_2$, respectively.

Table 1
Mechanical properties of sintered samples.

Sample	Relative density (%)	Flexure strength (MPa)	Vicker's hardness (GPa)	Fracture toughness (MPa $m^{1/2}$)
$MoSi_2$	78.2	108.64	4.65	3.63
$MoSi_2$–SiC	91.5	251.29	7.86	8.17

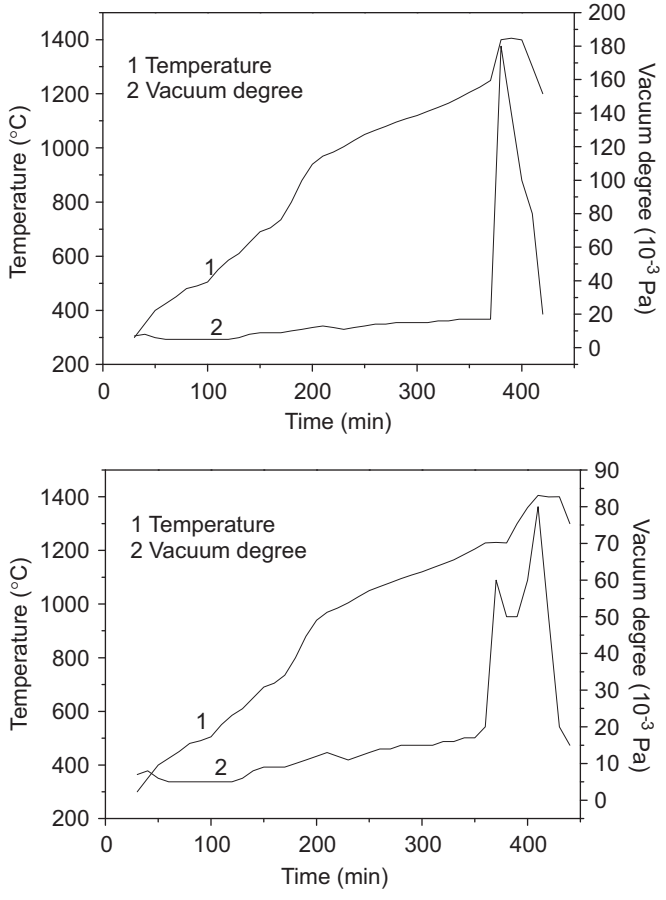

Fig. 3. changes of the temperature and vacuum degree during the microwave reaction sintering process: (a) normal process and (b) optimized process.

The heating procedure is very important to the densification process of the $MoSi_2$–SiC composite. At the beginning, we heated the reactant directly to 1400 °C with a selected power. (Fig. 3a). The temperature increased steadily below 1249 °C. Then, the temperature was suddenly out of control and jumped to 1400 °C from 1249 °C in 10 min, and the gas pressure of this microwave oven increased from 8.8×10^{-3} Pa to1.8×10^{-1} Pa as well. It indicated a violent exothermic reaction between Mo and Si occurred at this point. The heat generated from the reaction also promoted the evaporation of the impurities and resulted in the increasing of the pressure. Furthermore, we found the product had broken up into a few pieces with lots of cracks. To avoid the violent reaction, the temperature was then hold at 1230 °C for 20 min by controlling the heating power carefully. The reaction between Mo and Si powders proceeded smoothly as we expected (Fig. 3b). Later on, we heated the sample to 1400 °C for final densification procedure, and the desired dense $MoSi_2$–SiC composites were finally obtained.

4. Conclusions

A new method to prepare $MoSi_2$–SiC composite by the microwave reaction sintering from the element reactants has been developed. During the reaction, it is important to control the heating procedure precisely to obtain the high density product. $MoSi_2$–SiC composite also has very good mechanical properties. The relatetive density, flexural strength, Vicker's hardness and fracture toughness of 20% $SiC/MoSi_2$ are 91.5%, 251.29 MPa, 7.86 GPa and 8.17 MPa $m^{1/2}$, respectively.

Acknowledgment

This work was sponsored by Aid program for Science and Technology Innovative Research Team in Higher Educational Institutions of Hunan Province.

References

[1] A.D. Shan, W. Fang, H. Hashimoto, Y.H. Park, Effect of Mg addition on the microstructure and mechanical properties of $MoSi_2$ Alloys, Scripta Materialia 46 (2002) 645–648.
[2] J.Y. Gao, W. Jiang, Effect of La_2O_3 addition to modification of grain-boundary phase in $MoSi_2$, Journal of Alloys and Compounds 476 (2009) 667–670.
[3] J.J. Petrovic, A.K. Vasudevan, Key developments in high temperature structural silicides, Materials Science Engineering A 261 (1999) 1–5.
[4] R. Mitra, Mechanical behaviour and oxidation resistance of structural silicides, International Materials Reviews 51 (2006) 13–62.
[5] J.J. Petrovic, Mechanical behavior of $MoSi_2$ and $MoSi_2$ composites, Materials Science Engineering A 192–193 (1995) 31–37.
[6] X.L. Zhang, Z.L. Lu, Z.H. Jin, Electrical resistivity and microstructure of pressureless reactive sintered $MoSi_2$–SiC composite, Materials Chemistry and Physics 86 (2004) 16–20.
[7] D.Y. Oh, H.C. Kim, J.K. Yoon, I.J. Shon, One step synthesis of dense $MoSi_2$–SiC composite by high-frequency induction heated

combustion and its mechanical properties, Journal of Alloys and Compounds 395 (2005) 174–180.
[8] S. Yang, N. Chen, W.J. Liu, M. Zhang, In situ formation of $MoSi_2$/SiC composite coating on pure Al by laser cladding, Materials Letters 57 (2003) 3412–3416.
[9] M. Patel, J. Subramanyam, V.V. Bhanu Prasad, Synthesis and mechanical properties of nanocrystalline $MoSi_2$–SiC composite, Scripta Materialia 58 (2008) 211–214.
[10] J.I. Lee, N.L. Hecht, T.I. Mah, In Situ Processing and Properties of SiC/$MoSi_2$ Nanocomposites, Journal of the American Ceramic Society 81 (1998) 421–424.
[11] Q.D. Hu, P. Luo, Y.W. Yan, Microstructures and densification of $MoSi_2$–SiC composite by field-activated and pressure-assisted combustion synthesis, Journal of Alloys and Compounds 468 (2009) 136–142.
[12] P.C. Kang, G.Q. Chen, G.H. Wu, Synthesis nano-SiCp/$MoSi_2$ composites by in situ reaction sintering and low temperature oxidation behavior, Advances in Materials Research 105–106 (2010) 150–153.
[13] J. Pan, M.K. Surappa, R.A. Saravanan, B.W. Liu, D.M. Yang, Fabrication and characterization of SiC/$MoSi_2$ composites, Materials Science Engineering A 244 (1998) 191–198.
[14] F. Chen, J.G. Xu, Z.F. Hou, In situ pressureless sintering of SiC/$MoSi_2$ composites, Ceram. Int. http://dx.doi.org/10.1016/j.ceramint.2011.11.046.
[15] K. Peng, M.Z. Yi, L.P. Ran, Y.C. Ge, Reactive hot pressing of SiC/$MoSi_2$ nanocomposites, Journal of the American Ceramic Society 90 (2007) 3708–3711.
[16] R. Mitra, Y.R. Mahajan, N. Eswara Prasad, W.A. Chiou, C. Ganguly, Reaction hot pressing and characterization of $MoSi_2$/SiC_p composites, Key Engineering Materials 108–110 (1995) 11–14.
[17] R.D. Peelamedu, R. Rustum, D.K. Agrawal, Microwave-induced reaction sintering of $NiAl_2O_4$, Materials Letters 55 (2002) 234–240.
[18] I. Ganesh, R. Johnson, G.V.N. Rao, Y.R. Mahajan, S.S. Madavendra, B.M. Reddy, Microwave-assisted combustion synthesis of nanocrystalline $MgAl_2O_4$ spinel powder, Ceramics International 31 (2005) 67–74.
[19] M. Panneerselvam, A. Agrawal, K.J. Rao, Microwave sintering of $MoSi_2$–SiC composites, Materials Science Engineering A 356 (2003) 267–273.
[20] G.R. Anstis, P. Chantikul, B.R. Lawn, D.B. Marshall, A critical evaluation of indentation techniques for measuring fracture toughness: I, direct crack measurements, Journal of the American Ceramic Society 64 (1981) 533–538.