



Contents lists available at ScienceDirect

Int. Journal of Refractory Metals and Hard Materials

journal homepage: www.elsevier.com/locate/IJRMHM

Synthesis of Mo-Si-B intermetallic compounds with continuous α -Mo matrix by pulverization of ingot and hydrogen reduction of MoO_3 powders

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ARTICLE INFO

Article history:

Received 1 August 2016

Received in revised form 23 August 2016

Accepted 10 September 2016

Available online xxxx

Keywords:

Mo matrix

Mo-Si-B intermetallics

Pulverization

Hydrogen reduction

Microstructure

ABSTRACT

The fabrication of the intermetallic phase T2-Mo₃Si with continuous matrix of α -Mo was attempted with the combination process of high energy ball milling, pulverization of arc-melted ingot, addition of Mo by hydrogen reduction of MoO₃ and spark plasma sintering processes. High energy ball milling or arc melting of Mo-16.7Si-16.7B (at %) powders were performed to obtain to intermetallic phase T2 and Mo₃Si. The Mo phase of 57 vol% distributed intermetallic compound powders were prepared by hydrogen reduction of MoO₃ and further mixing of elemental Mo powders. X-ray diffractometry analysis revealed that the intermetallic phase T2-Mo₃Si can be produced by the pulverization process of arc-melted ingot. Hydrogen reduction of 1 vol% MoO₃ mixed intermetallic powder followed by further addition of Mo powders was a more adequate method enabling the homogeneous distribution of the Mo phase than that of added MoO₃ powders with total amount. The powder mixture was successfully consolidated by spark plasma sintering yielding a sound microstructure comprising the intermetallic phase T2-Mo₃Si uniformly distributed in a continuous matrix of α -Mo.

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1. Introduction

As a refractory metal, molybdenum (Mo) exhibits a high melting temperatures and excellent high temperature strength. However, pure Mo is easily oxidized above 500 °C by the formation of volatile MoO₃. Thus, the intermetallic phases Mo₅SiB₂ (T2) and Mo₃Si are of interest because of excellent high temperature strength and creep resistance by complex tetragonal structure as well as good oxidation resistance by the formation of a protective silicate glass [1–3]. However, their applications as high temperature structural parts in rocket, aircraft engines and gas turbines are restricted due to poor workability because of their low fracture toughness [4,5].

The fracture toughness of the intermetallic phases may be improved by the presence of the more ductile Mo phase. In particular, a typical microstructure of Mo-rich Mo-Si-B alloys yields a matrix of the Mo solid solution (α -Mo) providing good fracture toughness and ductility below 600 °C, and embedded intermetallic phases T2 and Mo₃Si for enhancing both creep and oxidation resistance [6,7]. However, in order to maximize the contribution of the ductile phase to the fracture

toughness and to achieve good oxidation resistance, it is a prerequisite for the α -Mo to be continuously distributed in fine intermetallic phases.

A variety of methods such as casting or powder metallurgy process have been suggested for producing Mo-rich Mo-Si-B alloys [8,9]. However, the fabrication of Mo-Si-B alloy with a required microstructure consisting α -Mo, T2 and Mo₃Si phases is quite a challenge because of high melting temperature of the intermetallic compounds and difficulty in achieving a homogeneous distribution of α -Mo phase. Recently, to overcome the difficulty in microstructure control, a mechano-chemical route using MoO₃, Si₃N₄ and BN powders has been suggested [10,11]. The prepared Mo-Si-B alloy showed a continuous α -Mo phase uniformly embedded by intermetallic phases and improved fracture toughness. However, the reported method has a difficulty in mass production because of need to exact reaction control between the initial powders.

In the present study, we have attempted to fabricate the α -Mo to be continuously distributed in fine intermetallic phases. The T2-Mo₃Si powders were prepared by high energy ball milling or pulverization of arc-melted ingot. The intermetallic compound powders with homogeneously coated α -Mo on their surfaces were synthesized by hydrogen reduction of MoO₃ powders and consolidated by spark plasma sintering for high relative density [12]. Microstructural characteristics of Mo-Si-B alloy powders and sintered composites with different fabrication

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method were analyzed by XRD and SEM. Also, the relationship between synthesis processing and microstructural characteristics was discussed.

2. Material and methods

The elemental powders of Mo (99.95%), Si (99.99%) and B (99.9%) were used as starting materials. The composition of the powder mixture was Mo-16.7Si-16.7B (at %) to obtain intermetallic phase T2 and Mo₃Si reported in Mo-Si-B phase diagram [13]. Two different fabrication processes were applied to obtain intermetallic compound powders. The first one was the mechanical alloying process, in which the elemental powders were ball milled in a high energy mill at durations up to 20 h using a ball to powder weight ratio of 15:1. The ball and jar were made of stainless steel. In the second method, elemental materials were arc-melted under a partial pressure of Ar, and they were then casted into water cooled molds. Finally the cast ingots were pulverized in a ball mill.

To fabricate the intermetallic compound powders with homogeneous distribution of α -Mo powders, the MoO₃ was mixed with prepared T2-Mo₃Si powders and hydrogen-reduced at 750 °C for 3 h. For comparison, 1 vol% of MoO₃ was mixed with T2-Mo₃Si powders and hydrogen-reduced in the same conditions. Then, elemental Mo powder was added to hydrogen-reduced powder mixtures. In all synthesized powders, the volume fractions of α -Mo and T2-Mo₃Si were 57 and 43%, respectively.

The α -Mo distributed intermetallic compound powders were sintered at 1200 and 1300 °C for 30 min in vacuum under a pressure of 30 MPa using spark plasma sintering (SPS, Sumitomo Coal Mining, Japan). Phase identification of the samples was performed by X-ray diffraction (XRD, Rigaku Denki Co., Japan) analysis, using Cu K α radiation. The morphology of the intermetallic compound powder and microstructure of the sintered body were examined using field-emission scanning electron microscopy (FE-SEM, JEOL Techniques, Japan) equipped with an energy dispersive X-ray spectroscopy (EDX).

3. Results and discussion

Mechanical alloying of Mo-16.7Si-16.7B powders was carried out to fabricate intermetallic phases. The XRD profiles registered for the ball-milled powders are shown in Fig. 1. The diffraction lines corresponding to intermetallic compounds of Mo₅SiB₂, MoSi₂ and Mo₂B as well as to elemental Mo can be observed in the powders milled for 10 h (Fig. 1a). It is clear from Fig. 1a that the initial powders were not completely converted into intermetallic phases after milling for 10 h, so it was decided to increase the milling time. However, powders ball-milled for 20 h shown in Fig. 1(b) have the main peaks in the spectrum corresponding to pure Mo and intermetallic phases, and the peaks Fe and Fe alloys are

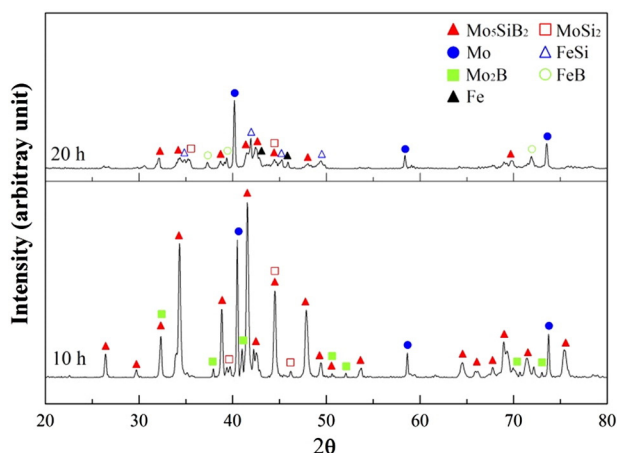


Fig. 1. XRD patterns for Mo-Si-B powders, ball-milled for 10 and 20 h.

also visible as minor phases. Existence of Fe peaks is mainly attributed to the contamination from the milling container and balls. To synthesize the T2-Mo₃Si phase without any reaction phases, the arc melting and pulverization processes were introduced.

Fig. 2(a) shows the XRD pattern of the arc-melted and pulverized powders. The Mo-16.7Si-16.7B powders were completely converted to T2 and Mo₃Si phase. Neither residual elemental metals nor reaction phase was observed. The pulverized intermetallic phases exhibit irregular particle shape shown in Fig. 3(a), and their average particle size was measured as 10.76 μ m. These results indicated that the intermetallic phases T2 and Mo₃Si can be fabricated more effectively by using arc melting and pulverization than by ball milling.

In order to fabricate the intermetallic compound powders with homogeneously coated α -Mo particles on their surfaces, the powder mixture of MoO₃ and T2-Mo₃Si was hydrogen-reduced at 750 °C for 3 h. The XRD profile registered for the powder mixture is shown in Fig. 2(b). After reduction in hydrogen atmosphere, the powder mixture was composed of Mo and T2-Mo₃Si as well as MoO₂ as unreduced phase. Also, platelet shape particles indicated by arrows were observed in reduced powder mixtures, as shown in Fig. 3(b). It is reported that reduction of MoO₃ by hydrogen takes place in two stages; MoO₃ \rightarrow MoO₂ at maximum reaction temperature range of 520–620 °C and MoO₂ \rightarrow Mo at 670–695 °C, and the MoO₃ is converted to MoO₂ with platelet shape via chemical vapor transport [14,15]. Thus, considering that the reduction temperature in this study was relatively high to reduce the MoO₃, the reason for the existence of MoO₂ phase can be explained by the insufficient reduction due to large amount of MoO₃ powders.

Fig. 2(c) and Fig. 4 show the XRD pattern and SEM images for the powder mixture of Fig. 3(b) after spark plasma sintering at 1200 °C for 30 min. As expected in XRD result of powder mixture, the lines for MoO₂ are found in XRD profile for sintered specimen. Also, inhomogeneous phases and large pores (dark) were observed in the sintered specimen, as shown in Fig. 4. These microstructural characteristics are mainly originated from the presence of unreduced MoO₂.

Thus, new approach to fabrication of powder mixture with fully reduced Mo was introduced by two step addition of Mo. Firstly, 1 vol% of MoO₃ powder mixed with pulverized T2-Mo₃Si powder were hydrogen-reduced at 750 °C for 3 h. Then, elemental Mo powder of the final

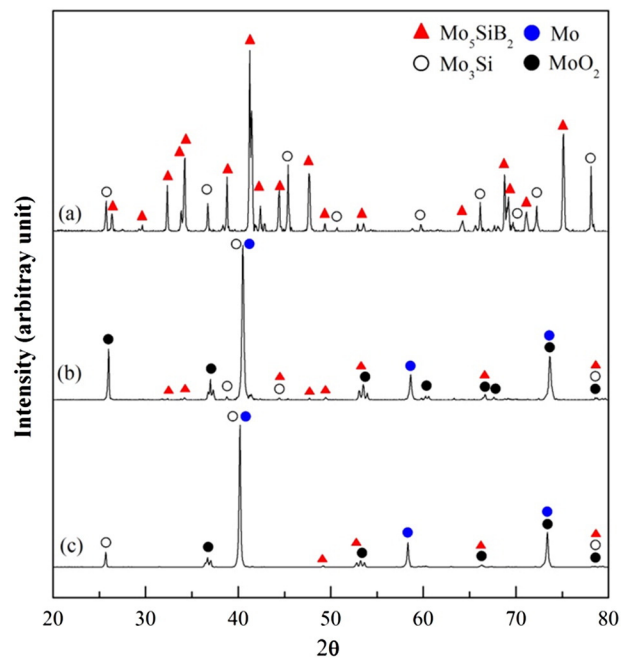


Fig. 2. XRD profiles of the Mo-Si-B powders at different stages of processing; (a) after arc melting and pulverization, (b) after MoO₃ addition and hydrogen reduction and (c) after spark plasma sintering at 1200 °C.

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