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Short communication

Effect of temperature and soaking time on the synthesis of $Mo(Al,Si)_2$

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Abstract

$Mo(Al,Si)_2$ has been prepared by hot pressing elemental powders of Mo, Al and Si, at 1200–1700°C and 22 MPa. Mo_5Si_3 or Mo_5Si_3C have been observed as minor phases in all the samples with 1300°C being the best processing temperature for synthesizing $Mo(AlSi)_2$: with minimum amount of impurities. As the soaking time increases, the amount of impurity phases also increases.

Keywords: Synthesis; $Mo(Al,Si)_2$

1. Introduction

$MoSi_2$ is a good high temperature structural material for use in aggressive environments. This stems from its high melting point (2030°C), excellent high temperature oxidation and corrosion resistance and moderate density (6.31 g/cc). However, $MoSi_2$, has some disadvantages like brittleness at temperatures lower than 1000°C, and poor creep resistance at temperatures above 1250°C, and it undergoes pesting at intermediate temperatures (300–600°C) [1–3]. It has been reported that aluminium addition to $MoSi_2$ and thus formation of $Mo(Al,Si)_2$ could improve the mechanical and oxidation properties of $MoSi_2$ [1]. $MoSi_2$ has a Cllb-type body centered tetragonal structure at temperatures lower than 1900°C and a C40 type hexagonal structure at temperatures higher than 1900°C [4]. $Mo(Al,Si)_2$ has a C40 type hexagonal structure at all temperatures. Therefore, $Mo(Al,Si)_2$ can be considered to be a high temperature hexagonal modification of $MoSi_2$, stabilized by the partial substitution of aluminium for silicon. Indeed, the improved mechanical properties of $Mo(Al,Si)_2$ are attributed to its more symmetric C40 hexagonal structure. The pesting of $MoSi_2$ at lower temperatures is also suppressed by aluminium addition, the volume change in the oxidation of $Mo(Al,Si)_2$ being much smaller (+4.9 vol.%) than for $MoSi_2$ (+85.6 vol.%). $Mo(Al,Si)_2$ prepared by micropyretic synthesis has some improved properties viz. higher fracture toughness, higher oxidation resistance etc. [4]. $Mo(Al,Si)_2$ has been synthesised to a good density by several methods viz., SHS with Mo, Al and Si [5], reactive hot pressing of $MoSi_2$, Mo and Al powders [6] and arc melting [1,2]. However, all these materials contained secondary unwanted phases and impurities. This work accounts for the preparation of high purity $Mo(Al,Si)_2$ by the hot pressing of elemental powders. From reactive infiltration of aluminium into $MoSi_2$ it was found that 17 at.% of Al reacted with $MoSi_2$ irrespective of the temperature and duration of infiltration [7]. Hence, the percentage chosen for aluminium was 17 at.% which also corresponds to the most stable form of $Mo(Al,Si)_2$ as evident from the phase diagram [8]. Hot pressing temperatures and soaking times were varied in an attempt to get $Mo(Al,Si)_2$ with the least amount of impurity phases.

2. Experimental procedure

Commercially available 99.5% pure Mo, Al and Si powders were mixed in stoichiometric proportion to obtain $Mo_{33.3}Al_{17}Si_{49.6}$. Pellets were prepared by hot pressing batches of 1 .5 gm of the mixture at 22 MPa in argon atmosphere at 1050–1700°C. Synthesis was also carried out at a constant temperature of 1300°C and different sintering times. All the specimens were then

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carefully polished for X-ray (D/Max 2200 Ultima X-ray Diffractometer) and microstructure analysis (LEO 4401).

3. Results and discussion

X-ray diffraction patterns of samples synthesized at different temperatures show $Mo(Al,Si)_2$ as the major phase (Table 1). In addition, samples prepared at temperatures lower than 1300°C contained Mo_5Si_3, Mo and Si as impurities. Samples prepared at 1500°C contained Mo_5Si_3, Mo_5Si_3C and Si as impurities. Above 1500°C i.e. at 1600 and 1700°C samples contain only Mo_5Si_3 and Mo_5Si_3C as impurities. Thus, samples prepared between 1300 and 1400°C contain the lowest amount of impurities in the form of Mo_5Si_3 though Mo_5Si_3 peaks at 1400°C are more intense than at 1300°C. A scanning electron micrograph of $Mo(Al,Si)_2$ prepared at 1300°C is shown in Fig. 1. $Mo(Al,Si)_2$ appears as dark grey particles and the aluminium content as calculated from the EDX analysis of the grey particles is in the range of 12–18 at.%.

At lower temperatures (<1300°C, i.e. 1050–1200°C) reactions may be incomplete. Hence, along with $Mo(Al,Si)_2$ there remain some unreacted Mo, Al and Si. Aluminium may be present as Al_2O_3. On the other hand, at 1500°C $Mo(Al,Si)_2$ is unstable and starts decomposing as follows:

$$7MoAl_{0.5}Si_{1.5} \rightarrow Mo_5Si_3 + 3.5Al + 2Mo + 7.5Si \quad (1)$$

At these temperatures the Mo_5Si_3 reacts with C (from the graphite die) to form Mo_5Si_3C. So at 1500°C there are impurities like Mo_5Si_3, Mo_5Si_3C and Si. With further increase in temperature i.e. at 1600 and 1700°C, there exists $Mo(Al,Si)_2$ along with some impurities of Mo_5Si_3 and Mo_5Si_3C. Thus, in all the samples prepared at different temperatures we can find that $Mo(Al,Si)_2$ is the major phase and Mo_5Si_3 is present as an impurity phase. The phase diagram of the Mo–Al–Si system [4] shows that $Mo(Al,Si)_2$ is stable only over a small compositional range. Hence, a little variation in composition leads to the formation of Mo_5Si_3 as an impurity phase.

Samples prepared at 1300°C proved to contain the lowest amount of Mo_5Si_3. Consequently, in another set of experiments $Mo(Al,Si)_2$ was prepared at a constant temperature of 1300°C and varying the hot pressing time. Fig. 2 shows the X-ray diffraction patterns of three samples prepared at 1300°C with soaking times of 15, 35 and 45 min. The X-ray diffraction peak corresponding to Mo_5Si_3 at 2θ of 42.65° starts becoming more intense with increasing soaking time, as given in Table 2. The less intense peaks (2θ: 62.0°, 67.7°, 70.1°, etc.) of Mo_5Si_3 start appearing as shoulders in the X-ray diffraction pattern of 35 min sintered sample giving an asymmetric look to the $Mo(Al,Si)_2$ peaks. In 45 min sintered sample these Mo_5Si_3 peaks increase in intensity and grow into

Table 1

Temperature (°C)	Phases present
1200	$Mo(Al,Si)_2$, Mo_5Si_3, Mo,Si
1300	$Mo(Al,Si)_2$, Mo_5Si_3
1400	$Mo(Al,Si)_2$, Mo_5Si_3
1500	$Mo(Al,Si)_2$, Mo_5Si_3, $Mo_5Si_3C_3$, Si
1600	$Mo(Al,Si)_2$, Mo_5Si_3, Mo_5Si_3C
1700	$Mo(Al,Si)_2$, Mo_5Si_3, Mo_5Si_3C

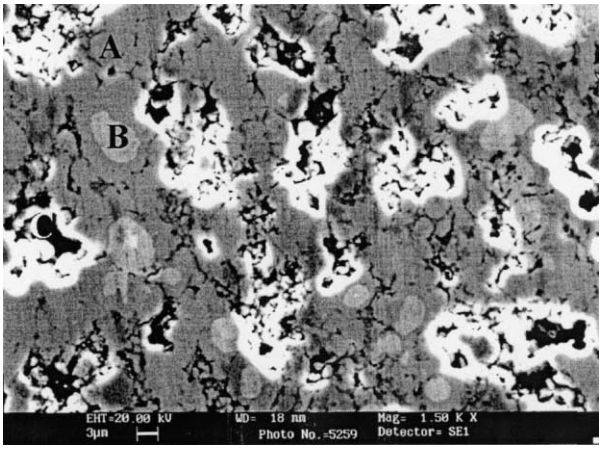

Fig. 1. Scanning electron micrograph of $Mo(Al,Si)_2$ prepared at 1300°C. $Mo(Al,Si)_2$ appears as dark grey particles (A), lighter coloured spots are due to Mo_5Si_3 (B) and the black regions correspond to the pores (C).

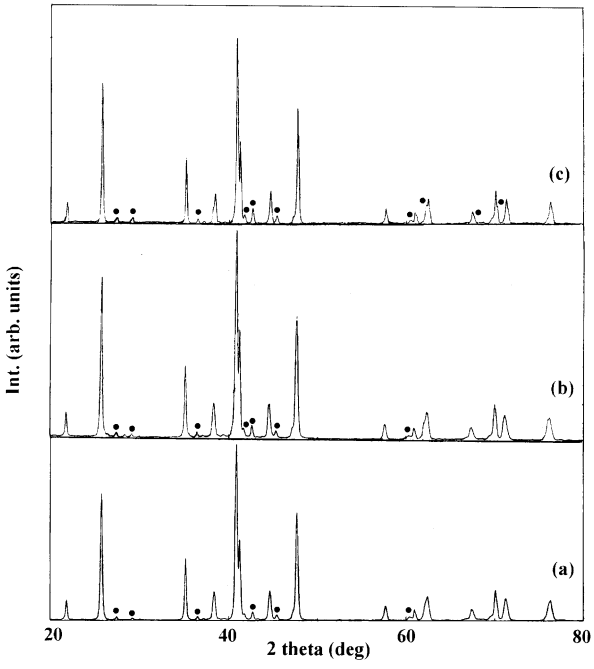

Fig. 2. X-ray diffraction pattern of $Mo(Al,Si)_2$ prepared at 1300°C with soaking time of (a) 15, (b) 35 and (c) 45 min; ●, Mo_5Si_3.

Table 2

Temperature (°C)	Soaking time (min)	I_1/I_2[a]
1300	15	0.049
1300	35	0.063
1300	45	0.081

[a] I_1, Intensity of Mo_5Si_3 peak; I_2, intensity of $Mo(Al,Si)_2$ peak.

clear peaks. It seems that $Mo(Al,Si)_2$ is not very stable at these temperatures for prolonged times. The concentration of Mo_5Si_3 increases because $Mo(Al,Si)_2$ which has already formed starts decomposing if it is kept at these temperatures for long periods of time.

4. Conclusions

$Mo(Al,Si)_2$ can be prepared with less than 10% impurity content by hot pressing elemental Mo,Si and Al at a temperature of 1300°C for 15 min. Small variations in temperature or sintering time lead to an increase in impurities. The main impurity phase is Mo_5Si_3.

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