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Preparation of $Mo(Si,Al)_2$ feedstock used for air plasma spraying

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Abstract: In order to prepare high quality $Mo(Si,Al)_2$ feedstock characterized with C40 phase, higher Al doping amount and excellent flowability, $Mo(Si_{1-x},Al_x)_2$ with different Al contents (x=0, 0.1, 0.2, 0.3, 0.4, 0.5) were synthesized by self-propagating high-temperature synthesis first and $Mo(Si_{0.6},Al_{0.4})_2$ was confirmed as the suitable material through X-ray diffraction analysis. A series of tests with different parameters of induction plasma spheroidization were applied to improving the flowability of feedstock. $Mo(Si,Al)_2$ feedstock with excellent flowability (26.2 s/50 g) was prepared through adding hydrogen into sheath gas and decreasing the powder feeding rate. The composition segregation occurred in the spheroidized powder after Al consumption and oxidation. The inhomogeneous structure of the same particle was caused by the asymmetric heating and cooling when particle passed through the plasma jet.

Key words: $Mo(Si, Al)_2$; C40 phase; self-propagating high-temperature synthesis; induction plasma spheroidization; composition segregation

1 Introduction

Molybdenum disilicide ($MoSi_2$) is an attractive coating material used in an oxidative atmosphere at high temperature due to the formation of an adherent and continuous SiO_2 film on its surface, which protects the material from further oxidation [1−3]. Whereas, there are several detrimental habits of $MoSi_2$ coating including the low ductility at ambient temperature, the pest oxidation in the intermediate temperature range, the evaporation of protective SiO_2 scale and the scale deterioration in the atmosphere containing water-vapor [4−6]. The former researches indicated that alloying additions to $MoSi_2$ can alter its anti-oxidation properties [7,8]. $Mo(Si, Al)_2$, with hexagonal C40-type structure, is one of the promising candidates for oxidation-resistance coating material. This material exhibits a good oxidation resistance at high temperatures over 1200 °C providing a continuous alumina scale and an Al−Si−O film at lower temperatures [9]. Besides, Al_2O_3 has proved to be effective for adjusting viscosity and improving the crystallization temperature of SiO_2 [10]. Therefore, $Mo(Si, Al)_2$ is considered to have definite advantages over $MoSi_2$ for oxidation-resistance coating at service temperatures up to 1400 °C [11].

Depending on Al contents, $Mo(Si, Al)_2$ possesses different crystal structures ($C11_b$, C40, C54) which show different oxidation behaviors and mechanisms [5,12]. Little discrepancy concerning critical value of crystal transformation occurs between the theory and experiment [13,14]. The $Mo(Si, Al)_2$ synthesis methods and operating condition may influence this critical value. In order to synthesize $Mo(Si, Al)_2$ with pure crystal structure, it is essential to clarify primarily the critical value of synthesis method and operating condition used in this work. Besides, although different preparation methods were applied to fabricating $MoSi_2$-based coatings, such as supersonic plasma spraying [15], high velocity oxygen fuel spraying (HVOF) [16], electro-thermal explosion ultrahigh speed spraying (EEUSS) [17] and air plasma spraying (APS) [18]. Feedstocks of metals, alloys and ceramics for thermal spray applications have to meet several specifications. In order to ensure high spray efficiency and better coating properties, particle shape, size distribution, powder

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flowability and density are the important factors that need to be controlled [19]. Inductively coupled plasma (ICP) with high enthalpy content has played an increasing important role in a wide range of technological processes, such as particle densification and spheroidization, plasma spray deposition of materials. It is a truism that particle spheroidisation is one of the successful applications of ICP and plays a key role in substantial improvement of powder quality and fluidity [20].

For the purpose of clarifying the critical value of $Mo(Si, Al)_2$ crystal transformation in self-propagating high-temperature synthesis (SHS) and obtaining $Mo(Si, Al)_2$ powder with pure crystal structure, a series of $Mo(Si_{1-x},Al_x)_2$ powders with different Al contents (x=0, 0.1, 0.2, 0.3, 0.4, 0.5) were synthesized by SHS in the present work. Through optimizing the parameters of induction plasma spheroidization (IPS), $Mo(Si,Al)_2$ feedstock with excellent flowability and nearly pure C40 crystal structure was also prepared. The crystal transformation of $Mo(Si, Al)_2$ during SHS and IPS were discussed in detail.

2 Experimental

2.1 Synthesis of $Mo(Si, Al)_2$ powder by SHS

Commercialized Mo, Si and Al powders (Qinhuangdao Eno High-Tech Material Development Co., Ltd., China) with the grain size of ~3 μm and the chemical purity of 99.9% were used as raw materials for combustion synthesis. The mole ratio of powder mixtures was chosen according to $Mo(Si_{1-x}, Al_x)$ with x range of 0−0.5. At the first step, reactant mixtures with stoichiometric ratio were well-blended, dried at 100 °C for 24 h, cold-pressed to a compact brick-like sample. Combustion reactions were initiated at one end of each sample, using electrical arc under 99.9% pure argon atmosphere with wave propagating mode (Dalian Ke Mao experimental facilities Co., Ltd., China).

2.2 Preparation of $Mo(Si, Al)_2$ feedstock

The SHS synthesized powder was crashed mechanically and meshed. The coarse powder (d_{50}= 50 μm) was then treated by IPS to improve the flowability and apparent density. The plasma was generated by an induction-plasma torch (Model PL 35, TEKNA Plasma Systems, Sherbrooke, Quebec, Canada) in connection with a radio frequency (R.F.) power-supply of 3 MHz. The fine powder (d_{50}=3.9 μm) was agglomerated into spherical or near-spherical particles with several tens of microns by spray drying process. The spray-dried particle may be crushed and blocked the powder feeding nozzle during APS process due to the low bonding strength of hollow agglomerated particles. Therefore, IPS process was also employed to densify the agglomerated particles. The parameters of IPS are listed in Table 1.

2.3 Characterizations

The surface morphologies and cross-sectional microstructures of powder were examined with a scanning electron microscope (SEM, Philips S−4800, Hitachi Ltd., Yoshida-Cho, Totsuka-Ku, Yokohama, Japan), which was equipped with an Oxford Inca energy dispersive spectroscopy (EDS) for chemical analysis. The phase structure of powder was analyzed by X-ray diffraction (XRD, Cu K_α, X'pert PRO, PANalytical B.V., Almelo, Netherlands). The particle size distribution was measured by a laser particle size analyser (Mastersizer 2000, Malvern, British). The particle flowability was measured by a standard flowmeter (FT−102B, Ningbo Rooko instrument Co., Ltd., China).

3 Results and discussion

3.1 Characterization of SHS powder

Figure 1 shows the XRD patterns of SHS synthesized $Mo(Si_{1-x}, Al_x)_2$ powder with different contents of Al. Without the adding of Al (x=0), nearly pure tetragonal $MoSi_2$ ($C11_b$) was identified for the product. When x=0.1, the coexistence of $C11_b$ and C40 phases was observed in the SHS product. In the case of $0.2 \le x \le 0.4$, the main phase has fully transformed to C40 phase with three weak peaks of $C11_b$. As the content of

Table 1 Main parameters of IPS

Test run No.	Powder	Gas flow rate/($mL \cdot min^{-1}$)			Reactor pressure/kPa	Feeding rate/($g \cdot min^{-1}$)	Mass balance (before/after)/g
		Central gas (Ar)	Sheath gas (Ar)	Sheath gas (H_2)			
1	Crushed	18	70	0	75.834	75	100/80
2	Crushed	18	60	7	75.834	75	100/68
3	Crushed	18	60	7	75.834	50	100/64
4	Spray dried	18	60	7	75.834	75	100/63
5	Spray dried	18	60	7	75.834	50	100/60
6	Spray dried	18	60	7	75.834	15	100/53