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Original Research Paper

Reaction mechanisms, resultant microstructures and tensile properties of Al-based composites fabricated in situ from Al-SiO_2-Mg system

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ABSTRACT

Reaction mechanisms, microstructures and tensile properties of the aluminum matrix composites made from Al-SiO_2-Mg system were investigated. When the temperature increased from room temperature to around 761 K, Mg dissolved into Al to form Mg-Al alloy. As the temperature increased to about 850 K, the remaining Mg reacted with SiO_2 to form MgO, Mg_2Si and Si as expressed in step reaction I: $6Mg + 2SiO_2 \rightarrow 4MgO + Mg_2Si + Si$. Finally, with a further increase in temperature, the remaining SiO_2 reacted with Al to produce Al_2O_3 and Si, while MgO reacted with Al_2O_3 to form $MgAl_2O_4$ as expressed in step reaction II: $4Al + 3SiO_2 + 2MgO \rightarrow 2MgAl_2O_4 + 3Si$. The Si also dissolved into matrix Al to form Al-Si alloy. Accordingly, its reaction process consisted of two steps and their apparent activation energies were 218 kJ/mol and 192 kJ/mol, respectively. As compared to the composites prepared by Al-SiO_2 system, its density increased from 2.4 to 2.6 g/cm^3, and its tensile strength and elongation increased from 165 MPa and 3.95% to 187 MPa and 7.18%, respectively.

1. Introduction

Chemical reaction synthesis has emerged as a simple, yet effective, technique for attaining high quality composites materials from abundant raw materials. Using this method a variety of nanocomposites materials have been developed, which show excellent mechanical, electrical and thermal properties [1–3]. In recent years, considerable attention has been drawn to developing in situ aluminum matrix composite materials, due to their excellent properties such as high specific strength and good wear resistance compared to the matrix [4–7]. Among these materials, micro- and nano-scale particulates reinforced aluminum matrix composites are of particular interest, since they can be prepared through economically viable processes [8–12]. Many fabrication routes have been developed for those types of composite, including self-propagating high temperature synthesis (SHS), London Scandinavian metallurgical (LSM), exothermic dispersion (XD), contact reaction (CR), directed melt oxidation (DIMOX), vapor liquid synthesis (VLS), reactive metal penetration (RMP), reactive hot press (RHP), friction stir processing (FSP) and microwave synthesis (MWS) [6,13–16]. Notably, the XD technique has shown distinct advantage, for it can be used to produce the composites with ultrafine reinforcing particles (<1 μm) and even nanoparticles (<100 nm), whose volume fraction can be varied over a wide range. The reaction systems such as Al-TiO_2 [17–20], Al-TiO_2-B [21–23], Al-TiO_2-C [24–26], and Al-TiO_2-B_2O_3 [27–29] have been used to synthesize the *in-situ* AMCs.

Recently, Al-SiO_2 system was also used to manufacture *in-situ* aluminum matrix composites using the XD method [30]. Al reacted with SiO_2 to form Al_2O_3 and Si. However, most of Si developed into large-sized blocks and cracked under mechanical load, which lowered the mechanical properties of the composites. To reduce the amount and size of Si, elements having a high affinity with Si may be incorporated to the Al-SiO_2 system. In view of this, we added the C into the Al-SiO_2 system and found that with increasing the C/SiO_2 mole ratio from 0 to 1.0, the amount of large-sized block Si decreased and disappeared finally. And that the tensile strength and elongation rate of the composites increased from 215 MPa and 3% to 245 MPa and 8%, respectively [31].

In this work, Mg powders were added into the Al-SiO_2 system to reduce the amount of the large block Si and as such produce fine or even nano-scale Mg_2Si particles as reinforcement agents. The reaction pathways were examined by using differential scanning

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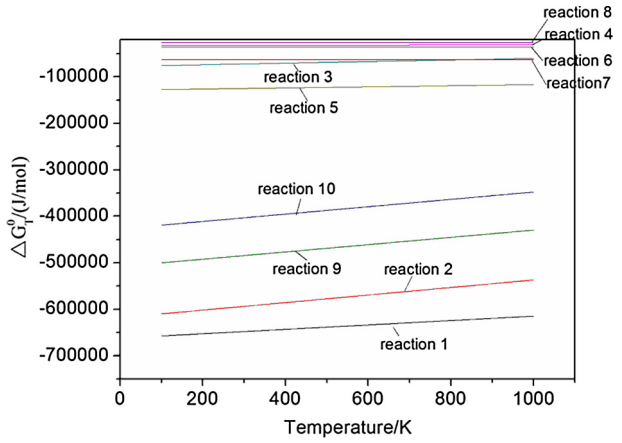

Fig. 1. Gibbs free energy of formation ΔG_T^0 of the potential reactions as a function of temperature for the Al-SiO_2-Mg system.

calorimetry (DSC), assisted by thermodynamic analysis. Moreover, the microstructure and mechanical properties of the resulting composites were determined, enabled by X-ray diffraction (XRD), Transmission electronic microscopy (TEM) and scanning electron microscopy (SEM) equipped with an energy dispersive analysis of X-rays (EDX).

2. Experimental procedures

2.1. Materials processing and microstructure

Al powders (purity 99.6%, particles size range ~50 μm, Shanghai Chemical Reagent Co., Ltd., China), SiO_2 powders (purity 99.0%, particles size range ~30 μm, Shanghai Chemical Reagent Co., Ltd., China) and Mg powders (purity 99.0%, particles size range ~100 μm, Shanghai Chemical Reagent Co., Ltd., China) were as raw materials used in the present study. A volume fraction of reinforcements of 30 vol% was predetermined in this study. As such, three powder ingredients were weighed to obtain a mixture of 76.4 wt % Al, 13.8 wt% SiO_2, and 9.8 wt% Mg, according to the following reaction: $4Al + 4SiO_2 + 8Mg \rightarrow 2MgAl_2O_4 + 3Mg_2Si + Si$. All the reaction products were expected to serve as reinforcements surrounded by the aluminum matrix, which is about 70 vol%. The powder mixture was ball-milled by a planetary ball milling (XQM-4, Nanjing University Instrument Co. Ltd., China) with the mass ratio of ball (size, 10 mm in diameter) to powder of 5:1 in a stainless steel jar (volume, 250 ml) under vacuum for 2 h. Then the mixed powders were in a drier (DHG-9030, Shanghai Sopo Instrument Co. Ltd., China) at temperature of 363 K for 120 min. After drying, the mixed powders were subsequently compacted into billets of 30 mm in diameter and 25 mm in height under a pressure of 150 MPa. The billets were heated in a furnace (VF1600, Bowen Instrument Co. Ltd., China) under vacuum up to 1373 K at a designed heating rate and held for 10 min in the furnace before cooling down to room temperature.

The microstructures of the resulting composites were observed using TEM (Tecnai $G^2$20 ST, FEI Co. Ltd. Hong Kong), SEM (Quanta 2000, FEI Co. Ltd., Holland) equipped with an energy dispersive analysis of X-rays (EDX). The densities of the composites were measured using the Archimedes principle.

2.2. Thermodynamic analysis

For indicating the reaction process of the Al-SiO_2-Mg system, the Al-SiO_2-Mg and its subsystems such as Al-Si, Al-Mg, Al-SiO_2 and SiO_2-Mg systems were prepared and tested by DSC analyzer (STA449C, NETZSCH Co. Ltd., Germany). Their mole coefficients are given as follows: 4Al-4SiO_2-8Mg, 28.7Al-Si, 4Al-8Mg, 4Al-4SiO_2 and 4SiO_2-8Mg, respectively. Small pieces of samples (each 5–10 mg) cut from the green billets were put into the DSC analyzer and were heated in argon atmosphere from the ambient (298 K) to 1373 K at a heating rate to test the temperatures at which the reactions would occur. In order to calculating the apparent activation energy, five different heating rates, i.e., 10 K/min, 15 K/min, 20 K/min, 25 K/min and 30 K/min were used.

2.3. Mechanical tests

The reacted billets were grinded to removed its surface and then hot extruded at ~723 K with an extrusion ratio of 10:1 to produce the rods of 7 mm in diameter, followed by cooling in air. The standard tensile test specimens (as per ASTM E8) were prepared by lathing and the tensile test were measured at room temperature using a universal materials testing machine (SANS/CMT5105, MTS Industrial Systems (China) Co., Ltd.) operating at a crosshead speed of 2 mm/min. All data were taken from at least three separated measurements. Tensile fracture surfaces were also observed using SEM and EDS to identify the damage mechanisms.

3. Results and discussion

3.1. Thermodynamic analysis

The Gibbs free energy of formation ΔG_T^0 can be calculated as $\Delta G_T^0 = \Delta H_{298}^0 - T\Delta S_{298}^0$ [32], where ΔH_{298}^0 is the standard enthalpy of formation and ΔS_{298}^0 the standard entropy of formation at ambient temperature, both of which can be derived from the following equations: $\Delta H_{298}^0 = \sum (n_i \Delta H_{i,f,298}^0)_{products} - \sum (n_i H_{i,f,298}^0)_{reactants}$ and $\Delta S_{298}^0 = \sum (n_i S_{i,298}^0)_{products} - \sum (n_i S_{i,298}^0)_{reactants}$, respectively. For the Al-SiO_2-Mg system, the potential reactions can be summarized as Eqs. (1)–(10) and their corresponding Gibbs free energies of formation ΔG_T^0 were determined based on the thermodynamic data [33] and using Eqs. (11)–(20):

$$6Mg + 2SiO_2 \rightarrow 4MgO + Mg_2Si + Si \quad (1)$$

$$4Al + 3SiO_2 \rightarrow 2Al_2O_3 + 3Si \quad (2)$$

$$2Mg + Si \rightarrow Mg_2Si \quad (3)$$

$$MgO + Al_2O_3 \rightarrow MgAl_2O_4 \quad (4)$$

$$3Mg + Al_2O_3 \rightarrow 3MgO + 2Al \quad (5)$$

$$MgO + SiO_2 \rightarrow MgSiO_3 \quad (6)$$

$$2MgO + SiO_2 \rightarrow Mg_2SiO_4 \quad (7)$$

$$MgSiO_3 + MgO \rightarrow Mg_2SiO_4 \quad (8)$$

$$4Al + 3MgSiO_3 \rightarrow 3MgO + 2Al_2O_3 + 3Si \quad (9)$$

$$4Al + 3Mg_2SiO_4 \rightarrow 6MgO + 2Al_2O_3 + 3Si \quad (10)$$

$$\Delta G_T^0 = -662,476 + 47.8T \quad (11)$$

$$\Delta G_T^0 = -617,877 + 80.5T \quad (12)$$

$$\Delta G_T^0 = -77,496 + 17.2T \quad (13)$$