Ceramics International 42 (2016) 9653–9659

Contents lists available at ScienceDirect

Ceramics International

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

Effects of oxide addition on the microstructure and mechanical properties of lamellar SiC scaffolds and Al–Si–Mg/SiC composites prepared by freeze casting and pressureless infiltration

Alateng Shaga, Ping Shen*, Rui-Fen Guo, Qi-Chuan Jiang

Key Laboratory of Automobile Materials (Ministry of Education), Department of Materials Science and Engineering, Jilin University, No. 5988 Renmin Street, Changchun 130025, PR China

ARTICLE INFO

Article history:
Received 21 October 2015
Received in revised form
3 February 2016
Accepted 7 March 2016
Available online 8 March 2016

Keywords:
Al–Si–Mg/SiC
Composites
Freeze casting
Pressureless infiltration
Compressive strength
Elastic modulus

ABSTRACT

Silicate-bonded porous SiC scaffolds with lamellar structures were prepared by freeze casting and liquid-phase sintering. It was found that the viscosity and solidification velocity of SiC water-based slurries with 30 vol% solid loading decreased with increasing Al_2O_3–MgO (AM) addition. As the AM content increased from 10 to 30 wt%, the lamellae of the sintered scaffolds became denser and the porosity decreased from $69 \pm 0.5\%$ to $62 \pm 0.5\%$, while the compressive strength improved from 25 ± 2 to 51 ± 2 MPa. The dynamics of pressureless infiltration for an Al–12 Si–10 Mg alloy on the SiC porous scaffold was measured and the composites with lamellar-interpenetrated structures were successfully produced. Both the compressive strength and the elastic modulus of the composites increased with increasing AM content. The maximum strength reached 952 ± 24 MPa and the highest elastic modulus about 156 GPa, respectively, in a longitudinal direction, increasing about 32% and 11% as compared with those of the composites without AM.

1. Introduction

Silicon carbide reinforced aluminum matrix composites have attracted great interest because of their improved properties such as high strength, better wear resistance, increased stiffness and enhanced creep resistance as compared to the monolithic aluminum alloy, and thus are widely used in aerospace, automotive and electronic packaging areas [1,2]. Increasing demands on such materials lead to a widespread research on the relationship between structure and property. Inspired by the fantastic microstructure and fabulous properties exhibited in natural materials such as nacre, the concept of biomimetic material design has been advocated as an innovative idea for decades [3]. Recently, freeze casting has received wide attention as a novel technique to produce lamellar porous ceramics with nacre-like architecture due to its simple operation, environmental friendliness and controllable pore size and porosity in the scaffolds [4,5]. Moreover, researchers have produced lamellar porous Al_2O_3 scaffolds by freeze casting aqueous alumina slurries and then fabricated shell-like composites by infiltrating an Al alloy into the scaffolds, in which hard and ductile phases were alternately arranged and both strength and toughness of the composites were significantly improved [6–9]. However, up to date, only very limited studies have focused on the preparation of laminated Al/SiC composites possibly because it is hard to sinter the SiC scaffolds due to the covalent nature of SiC and difficult to control the interfacial reaction between Al and SiC [10,11].

Conventionally, high temperatures over 2100 °C are needed for solid-phase sintering of the SiC ceramics, limiting their production [5]. The addition of oxides as sintering aids to the starting composition is effective in lowering the sintering temperature by formation of some intermediate bonding phases such as mullite ($Al_6Si_2O_{13}$), cordierite ($Mg_2Al_4Si_5O_{18}$), olivine (Mg_2SiO_4) and yttergranate ($Y_3Al_5O_{12}$) and thus could be used to produce the porous SiC scaffolds with reasonable strength [5,12–14]. These silicate phases may not only enhance the strength of the porous SiC scaffolds, avoiding their collapse during metal infiltration, but also act as a protective layer to avoid direct contact or at least reduce the reaction of SiC with molten Al for the formation of harmful Al_4C_3 phase. For example, Liu et al. [10] prepared the SiC scaffolds with the addition of 10 wt% (Al_2O_3+Y_2O_3) by freeze casting and liquid-phase sintering at 1900 °C for 1 h under a N_2 atmosphere. Subsequently, they produced the 2024 Al/SiC co-continuous composites by squeeze-casting of a 2024 Al alloy into the porous SiC scaffolds under a pressure of 80 MPa at 800 °C. They investigated the effects of freezing temperature and ceramic fraction

* Corresponding author.
E-mail address: shenping@jlu.edu.cn (P. Shen).

http://dx.doi.org/10.1016/j.ceramint.2016.03.052
0272-8842/

on the mechanical properties of the composites, without paying attention to the effect of the sintering additive. In our previous work [11], we prepared the novel lamellar-interpenetrated Al–Si–Mg/SiC composites by pressureless infiltration of an Al–12 Si–10 Mg alloy into the porous SiC scaffolds without AM. The highest compressive strength of 722 ± 35 MPa was achieved in the composites with 30 vol% SiC when loaded in the longitudinal direction parallel to the freezing.

The objective of the present work is to investigate the effects of oxide addition (Al_2O_3–MgO) on the microstructure and mechanical performance of the SiC scaffolds and the Al–Si–Mg/SiC composites, which were prepared by freeze-casting and pressureless infiltration, respectively. Meanwhile, the dynamics for infiltration of an Al–12 wt% Si–10 wt% Mg alloy into the silicate-bonded porous SiC scaffold was measured and their interfacial reaction was analyzed.

2. Experimental procedure

Commercial SiC (D_{50}=5 μm, 98.5% purity), Al_2O_3 (D_{50}=5 μm, 99.5% purity) and MgO powders (D_{50}=0.5 μm, 99.9% purity) were used as raw materials. 10–30 wt% Al_2O_3–MgO (AM) powders with a fixed weight ratio of 6:1 were added into the water-based slurry as oxide addition. Aqueous suspensions with a total of 30 vol% solid loading were prepared using 0.8 wt% carboxymethyl cellulose Na salt (CMC-Na) as dispersant at a pH value of 10 regulated by aqueous ammonia (NH_4OH). The slurries were then ball-milled for 12 h using alumina balls and de-aired by stirring in a vacuum desiccator for 20 min. Their viscosity and rheological properties were measured by a rheometer (DV−79+PRO, Nirun Co. Ltd., China).

After the measurement, the slurry was poured into a transparent polycarbonate mold and kept at −10 °C at bottom for single-side directional solidification by heat conduction through a Cu rod, whose end was immersed in liquid N_2. The freezing rate of the ice front was estimated by recording freezing time as a function of the distance away from the bottom. After frozen, the sample (⌀15 × 25 mm^2) was demolded and freeze-dried at −50 °C for 24 h under a 10 Pa vacuum. The dried samples were heated at 5 °C/min in air up to 1200 °C, holding for 1 h to achieve sufficient oxidation at SiC surfaces. Then, the furnace was evacuated to a vacuum and purged with Ar. Subsequently, the samples were heated in the flowing Ar atmosphere at 5 °C/min up to 1500 °C, holding for 2 h. Finally, they were cooled at a rate of 5 °C/min.

Next, pressureless infiltration for an Al–12 Si–10 Mg alloy on the porous scaffold was monitored using a charge-coupled-device (CCD) camera through quartz windows at both sides of the alumina tube. Firstly, the alloy was cut into sizes of 9 × 9 × 9 mm^3 and placed on the top of the SiC scaffold, and then the couple was held in an alumina crucible and placed into the center of the alumina tube. The tube was first evacuated to a vacuum less than 10 Pa and then purged with high-purity (99.999%) N_2. When it was heated to 600 °C at a rate of 5 °C/min in the flowing N_2 atmosphere (flow rate 3 L/min), photographs were taken by the CCD camera through the quartz window, which was illuminated using a 10 mW He–Ne laser, to record the change in the shape of the alloy. The tube furnace was further heated to 950 °C at a rate of 5 °C/min and then dwelled for a certain time. During this stage, the alloy melted and spontaneously infiltrated into the porous scaffold. The infiltration rate was estimated according to the time-dependent change in the height of the liquid alloy. The purpose of this measurement is just to determine the processing parameter (mainly holding time) for the subsequent spontaneous infiltration experiments.

The microstructures in the sintered scaffolds and the composites were observed using an optical microscope (Axio Imager A2m, Carl Zeiss, Germany) and a scanning electron microscope (SEM, Evo18, Carl Zeiss, Germany) equipped with an energy dispersive spectrometer (EDS). The phase constituents were determined by X-ray diffraction (XRD, D/Max 2500PC Rigaku, Japan). The density of the sintered scaffolds was calculated by measuring their dimensions and mass, and this value was further used to calculate porosity. Moreover, the SiC scaffolds in dimensions of ⌀15 × 25 mm^2 and the composite samples, which were sectioned from the upper part and cut into 5 × 5 × 10 mm^3 along the direction parallel to the freezing direction, were used for compression test at room temperature using a universal testing machine (Instron 5689, Instron Corp., USA) at a strain rate of 3×10^{-4} s^{-1}. Three specimens were tested to obtain average strength. The Al–12 Si–10 Mg alloy with the same dimensions was also measured under the same conditions to provide with contrastive reference.

The density of the composites was measured using Archimedes' method. To evaluate the elastic behavior of the processed materials, the composites for ultrasonic pulse echo measurement were cut into 10 × 10 × 10 mm^3 using a diamond saw. An ultrasonic thickness gauge (Olympus 38DL PLUS, USA) with a longitudinal wave probe (M112–RM, 10 MHz) and a shear wave probe (V156–RM, 5 MHz) was used to measure the velocities of longitudinal wave and shear wave that were propagated in the composites in the longitudinal (ice growth) direction. The values of longitudinal wave velocity, C_L, shear wave velocity, C_S, and density, ρ, were used to calculate elastic modulus, E, using a calculation method described in details in our previous study [11].

3. Results and discussion

3.1. Preparation of SiC scaffolds

Fig. 1 shows the relationship between the viscosity and shear rate of the slurries with the AM contents ranging from 0 to 30 wt%. It can be seen that the viscosity of all the slurries decreases with increasing shear rate, showing shear-thinning behavior. Moreover, the viscosity of the slurry decreases with increasing AM content, which is consistent with the result obtained by Zhang et al. [15] and can be attributed to the fact of a higher zeta-potential of Al_2O_3 than that of SiC in alkalinous condition, giving a larger repulsive force.

Fig. 2 shows the variation in solidification velocity with the distance away from the cooled bottom rod during freeze casting of the SiC slurries with different AM contents. Clearly, the

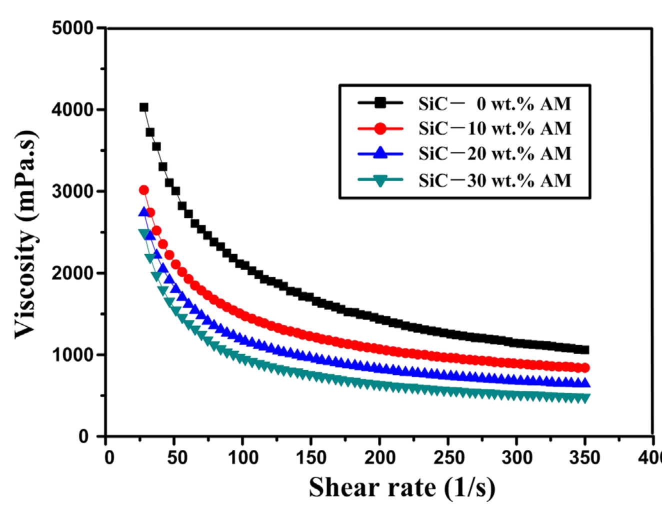

Fig. 1. Dependence of viscosity on shear rate for the SiC slurries with different AM contents.