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Fabrication of aluminum matrix composites by quick spontaneous infiltration process through combustion reaction of Al–Ti–B_4C–CuO powder mixtures in molten aluminum

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A novel, quick, spontaneous infiltration process that combines with a combustion reaction from a powder mixture of Al–Ti–B_4C–CuO is presented. The process is realized in a simple and economical way, with the whole process being performed in air in a few minutes. The microstructures of the composites exhibit quasi-continuous three-dimensional network structures composed of infiltrated aluminum and B_4C particles surrounded by the aggregates of the reaction products. The composites exhibit a high elastic modulus with a low coefficient of thermal expansion.

Keywords: Metal matrix composites; Liquid infiltration; Self-propagating high-temperature synthesis; Microstructure; Mechanical properties

Aluminum matrix composites with a high volume fraction of ceramic particulates such as SiC, TiC, B_4C and TiB_2 have high potential for advanced structural applications when the properties of specific strength, modulus, resistance to wear and thermal stability are at the required levels [1,2]. Several processing techniques have been developed to fabricate metal matrix composites (MMCs), such as stir casting, powder metallurgy and melt infiltration [3–6]. Among these, melt infiltration is a good way to produce MMCs with a high volume fraction of ceramic particulates.

In general, aluminum melt does not wet ceramics well and this gives rise to negative pressure. To overcome the wetting problem, several approaches, such as pressure infiltration [7,8], pressureless infiltration [9] and reactive infiltration [6], have been established to enhance the infiltration of molten metal into preforms. However, each fabrication technique has its own advantages and demerits. Applying pressure to the melt allows the negative capillary pressures to be overcome in a short process time, but the external pressure may damage the preforms and the process requires cumbersome pressing equipment. The pressureless infiltration process has several advantages over pressure infiltration; however, the cost remains high because the process requires a vacuum or an inert atmosphere, a relatively high processing temperature and a longer processing time compared to that of pressure infiltration. Reactive infiltration mainly relies on the in situ reaction to promote infiltration by coating ceramic powders and adding reactive elements to the matrix. While MMC products can be made easily in this way, with good near-net-shape characteristics, it has proved difficult to promote quick infiltration under these conditions.

B_4C is an attractive reinforcement for aluminum alloys due to its low density, high hardness, high elastic modulus and low coefficient of thermal expansion. However, due to its poor wettability [10] and interfacial reaction [11] with aluminum, B_4C is not always a good choice of reinforcement for MMCs. Hence, much effort has been made to enhance the wettability by coating B_4C particles with TiO_2 or adding the reactive element Ti so as to obtain desirable interfacial reaction products, such as TiB_2 and TiC [3,12–13].

In this investigation, we present the discovery of a novel quick spontaneous infiltration process that enables us to produce aluminum matrix composites containing B_4C with a sound microstructure. The core innovation

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of the process is that it combines a simple spontaneous infiltration in air with a combustion reaction of Al–Ti–B_4C–CuO in molten aluminum. This realizes quick infiltration within a few minutes, thus the process is promising for scaled-up production of composites in a simple and economical way.

The following powders were used as starting reagents: Ti (99.5%, 50 μm), Al (99.5%, ~30 μm), B_4C (99.5%, 16 μm), and CuO (99.95%, ~10 μm) powders. The reagent powders were mixed at a proper mole ratio (Ti:B_4C:Al:CuO = 3:4:6:0.4) for 1 h and then compacted at room temperature into a cylindrical shape to make a pellet ($h = 28$ mm, $d = 35$ mm, 55–60% of the theoretical density). The pellet was dried in a vacuum oven at 200 °C for 2 h to eliminate any moisture. Figure 1 provides a schematic representation of the process. The pellet was placed inside a graphite tube mold and the mold was put into a pure aluminum melt that was kept at 1173 K (Fig. 1a). Before ignition (preheating stage in Fig. 3a), the top surface of the pellet was exposed to air in order to remove the pellet's internal gas and moisture easily, which are inevitably absorbed in the surface of powders. Immediately after the onset of ignition, as evidenced by a dazzling light originating from the reaction of the powder mixtures (Fig. 1b; corresponding to the onset of ignition shown in Fig. 3a), the mold was dipped into the aluminum melt so that the aluminum melt would infiltrate into the pellet (Fig. 1c; corresponding to the reaction/infiltration stage shown in Fig. 3a). Finally, the mold was taken out from the melt, with the top surface coated by the melt, and cooled down in air (Fig. 1d; corresponding to the stabilization stage shown in Fig. 3a). The whole process was completed in 4 min in air.

For the microstructural analysis, samples were sectioned and prepared using standard polishing procedures, the final polish being with a 0.05 μm colloidal silica suspension. Microstructures were investigated by means of an optical microscope and a scanning electron microscope (SEM) equipped with an energy-dispersive spectrometer (EDS). X-ray diffraction (XRD) analysis with Cu K_α radiation was also carried out to identify the phases.

Figure 2 shows the overall aspect and the microstructures of the composites. The composites retained the initial shape of the pellet and were free of cracks; the aluminum melt was infiltrated up to the top surface of the pellet, as shown in Figure 2a.

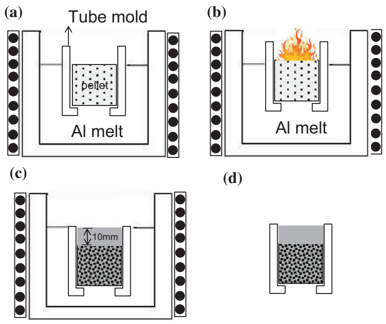

Figure 1. Schematic diagram of the processing procedures. (a) Exposing the top surface of pellet to air in the melt before ignition (preheating stage). (b) Onset of ignition. (c) Dipping the pellet fully into the melt (reaction/infiltration stage). (d) Cooling down of the pellet after removal from the melt.

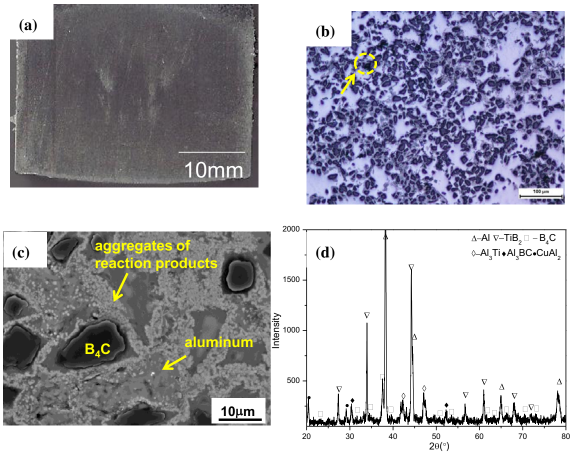

Figure 2. (a) A cross-section of the pellet composites infiltrated with Al upon combustion reaction and microstructures of the composites analyzed (b) with an optical microscope and (c) by backscattered electrons using a field emission gun-SEM; (d) XRD pattern showing constituent reaction products of the composites.

The microstructures of the composites shown in Figure 2b and c exhibited quasi-continuous three-dimensional network structures composed of infiltrated aluminum and B_4C particles surrounded by a large amount of aggregates of the reaction products throughout the specimens. It was confirmed by SEM/EDS and XRD analysis that the main reaction products are TiB_2 and Al_3BC (see Fig. 2c and d). In addition, small amounts of $CuAl_2$ and Al_3Ti were also observed in the microstructure. The aggregates of the reaction products, including TiB_2, Al_3BC and Al_3Ti, were confirmed by TEM to have very fine sizes (less than 0.5 μm in size) and to be disconnected from each other. The detailed results and related reaction mechanism are not presented here but will be reported in a future study.

In an Al–Ti–B_4C system with a mole ratio of Ti/B_4C = 0.75, the following reactions are expected:

$$3Ti + 4B_4C + 6Al = 2TiB_2 + TiC + 3B_4C + 6Al \qquad (1)$$

$$3Ti + 4B_4C + 6Al = 3TiB_2 + 2Al_3BC + 2B_4C \qquad (2)$$

In the microstructures, no trace of TiC is observed and a large amount of Al_3BC is formed instead; this means that the reactions of the system mainly follow Eq. (2) and that most of the Ti elements are consumed to form TiB_2.

Ignoring the content of CuO and considering the content of the aluminum melt that was infiltrated, the volume fraction of B_4C (16 μm in size) in the initial pellet is calculated to be 25.7%. The volume fractions of B_4C, TiB_2 and Al_3BC were estimated, according to Eq. (2), to be 13.4%, 14.3% and 22.5%, respectively. However, the measured volume fraction of B_4C in the composites was 18.3%, with an average particle size of 7.7 μm. Al_3Ti is also present in the microstructures, as shown in Figure 2d; this means that an incomplete reaction occurred, with fewer B_4C particles decomposed, according to the following equation: