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Synthesis and Characterization of Aluminium Base in situ Metal Matrix Composites by Spark Plasma Sintering

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Abstract

Fe-aluminide and alumina reinforced in-situ aluminium based metal matrix composite was prepared by spark plasma sintering (SPS) of aluminium and nanosized Fe_2O_3 powder mixture. In-situ reinforcements were formed during SPS by exothermal reaction between aluminium and nano-size Fe_2O_3 particle. The thermal characteristics of the in-situ reaction were studied by differential scanning calorimetry (DSC). Field Emission Scanning Electron Microscopy (FESEM) along with the Energy Dispersive Spectroscopy (EDS) and X-ray diffraction (XRD) techniques were used to study the microstructural architecture of the composites as a function of SPS temperature and the volume fraction of reinforcement. Microhardness measurement of the composite shows significant increase in hardness with increase in SPS temperature and volume fraction of secondary phase.

Keywords: In-situ, Fe-aluminide, metal-matrix-composite, nanosize, Spark plasma Sintering

1. Introduction

During the last two decades, metal matrix composites (MMC) have emerged as an important class of materials for structural, wear, thermal, transportation and electrical applications. These composites possess superior strength to weight and high strength to cost ratio, when compared with their equivalent commercial alloys (Tjong & Ma, 2000). It has been reported that the properties of the MMCs are influenced to a great extent by the nature of the reinforcements and their distribution in the host metal matrix (Tjong & Ma, 2000; Everett, & Arsenault, 1991; Ibrahim, Mohamed, & Lavernia, 1991). Traditionally, these composites were prepared by powder metallurgy and diverse melting and casting routes (Chawla, 2012; El Baradie, 1990). However, in all these cases the scale of reinforcing phases and their distribution is constrained by the starting size of the reinforcements (Sheibani & Najafabadi, 2007; Gotman, Koczak, & Shtessel, 1994).

Recently, the technique of reinforcing metal matrices by in-situ reaction has gained considerable attention (Kuruvilla, Prasad, Bhanuprasad, & Mahajan, 1990; Laksmi, Lu, & Gupta, 1998). In this technique, the reinforcing phase(s) is (are) formed in the host matrix via in-situ chemical reaction between the matrix and the precursor element(s)/compound(s) during the composite fabrication. These composites, termed as in-situ metal matrix composites and often referred as second generation metal matrix composites (Gu, & Shen, 2007; Song et al., 2008; Li et al., 2003; Tong, 1998), offer many advantages over the conventional composites. The most important advantage among many is that the reinforcements so formed by the in-situ reaction are finer in size and their distribution is more uniform, resulting in better mechanical properties of composites. However, here it would be worthwhile to mention that in most of the cases due to the high initiation temperature of the in-situ reaction(s), formation of the reinforcements within the host matrix necessitates high processing temperature. But processing the composites at high temperatures involves the risk of oxidation of the matrix and may also cause agglomeration and coarsening of the reinforcements, which will cast an adverse influence on the mechanical properties of the composite.

In the above perspective, it is to be noted that nano dimensional particles possess better chemical reactivity as compared to their coarser counterparts (Shen, Zou, Jin, & Jiang, 2007). Therefore, it is expected that the use of nanosized precursors in the processing of the in-situ composites will decrease the onset temperature and thereby would improve the kinetics of the in-situ reaction(s). Moreover, occurrence of such reactions at lower temperature reduces the chance of oxidation, agglomeration and coarsening of the reinforcements.

In recent years, the spark plasma sintering (SPS) process has been developed to obtain dense, near-net shaped bulk products from nanometer-size powders. During SPS, densification proceeds by applying high direct, pulsed current at low potential through the powder compact with externally applied pressure (Omori, 2000; Xü, Jia, & Cao, 2005; Nygren & Shen, 2003). In the process, the powder compact is subjected to cycles of very rapid heating and cooling with very short holding time at a sintering temperature, lower than that in normal hot pressing (Nygren & Shen, 2003). Recently, mechanical alloying followed by SPS has been used to fabricate Al_3Ti (Nygren & Shen, 2003) and Al_3Zr (Lee, Moon, & Lee, 2006; Moon, Kim, & Lee, 2002; Lee et al., 2003) based nanocrystalline, intermetallic alloys with attractive mechanical properties.

The present investigation aims at developing a mechanically stiffer and stronger in-situ Al matrix composite containing finely dispersed Al_2O_3 and Fe-aluminide reinforcements by spark plasma sintering of Al powder and nanosized Fe_2O_3 powder.

2. Experimental Procedures

2.1 Composites Fabrication

Aluminium powder of 99% purity with an average particle size ~20 μm (Loba Chemie Pvt Ltd, Mumbai) and nanosized Fe_2O_3 powder (average particle size ~ 5.6 nm) prepared by dry milling were used for preparing ($Al+Fe_2O_3$) green powder mix. The powders were weighed separately and mixed in a small plastic jar containing 10 number of 4 mm steel balls. The weight of aluminium and nanosized Fe_2O_3 powders was adjusted in such a way so that the composite formed by the complete in-situ reaction will contain the desired volume percent reinforcements as shown in Table 1.

Table 1. Composite sample identity with the expected vol. % of the reinforcing phase

Sample designation	Volume % of reinforcement
AF_1	10
AF_2	20
AF_3	30
AF_4	40
AF_5	50

The powder mix, so prepared were put into a graphite die (outside and inside diameters are 30 and 15 mm respectively) and consolidated by spark plasma sintering (Sumitomo Coal Mining, Japan). SPS was carried out into vacuum, at 700, 800 and 900 °C under 50 MPa uniaxial pressure and 10 minute holding time. The rate of heating was maintained at 50 K/min. The SPSed pellets were cylindrical 15 mm diameter and 5 mm thickness. To avoid any carbon contamination from graphite die, SPSed pellets were grounded by abrasive paper.

2.3 Characterization

The thermal characteristics of the in-situ reaction in the green compacts, expected to yield 100 vol.% (Al_2O_3 + Fe-aluminide) reinforcement on complete reaction, were studied under argon gas atmosphere with the aid of a differential scanning calorimeter (DSC, Mettler, TA400). The flow rate of argon gas was maintained at 80cc/min throughout the experiment. The green compacts were heated from room temperature to 700^0C at a predetermined heating rate of 10^0C/min and then cooled to room temperature at a cooling rate of 10^0C/min. In addition to the above studies, DSC investigation under identical environmental condition was also performed on the ball milled powder sample of Fe_2O_3 so that the results can be used as a reference to interpret the thermal characteristics of the compact during heating and cooling.

XRD studies on the SPS composite samples were carried out using Cu-K_α radiation. Prior to the analyses, the XRD patterns were corrected for the effects of the $K_{\alpha 2}$ radiation. The particle size of the ball milled Fe_2O_3 powders were evaluated from the XRD patterns by taking recourse to the single line profile analysis technique (Klug & Alexander, 1974).

A Field emission gun assisted scanning electron microscope (FESEM, Carl Zeiss, Supra 40) along with the energy dispersion spectroscopy (EDS) was used to study the microstructural architecture and composition of the different phases of the composite samples.

Average values of hardness on different phases of the composite samples were measured with the help of a standard Vickers hardness tester (Type 3212, Zwick, Ulm Germany) using a load of 50gm applied for 15s. The average hardness values were determined from 10 identical measurements.

3. Results and Discussion

3.1 Characteristics of in-situ Reaction

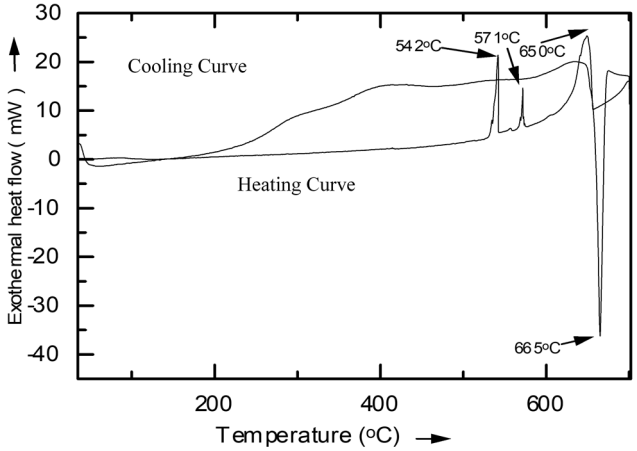

Figure 1. DSC heating and cooling plot of the green compact expected to yield 100vol% (Al_2O_3 + Fe-aluminide) reinforcement on complete reaction

Figure 1 presents the DSC results obtained with the green compact during the heating and cooling cycle without any isothermal holding at 700^0C. The exothermic peak observed at 650^0C in the cooling curve is attributed to the heat released during solidification of the unreacted aluminum. Here, it can be clearly seen that the total amount of heat evolved during solidification is much less compared to the heat absorbed during melting of aluminum. This suggests that the in-situ reaction between nanometric iron oxide particles and molten aluminum is more favorable than the solid-state reaction due to better and intimate contact of Fe_2O_3 particles with molten Al. The exothermic peak observed in the cooling curve at 571^0C may be assigned to either magnetic ordering or ($\alpha+\beta_2$) transformation of Fe_3Al. Similarly, the peak at 542^0C can be ascribed either to $\alpha+DO_3$ or entirely DO_3 transformation of Fe_3Al. Therefore, we conclude that the in-situ reaction between nanosize Fe_2O_3 and Al will lead to the evolution of Fe-aluminide particles in the Al matrix. However, it is to be noted that the evolution of intermetallic particulate reinforcement and their type would depend on the temperature and amount of aluminum and nanosized Fe_2O_3 powder.

3.2 Microstructure and Properties

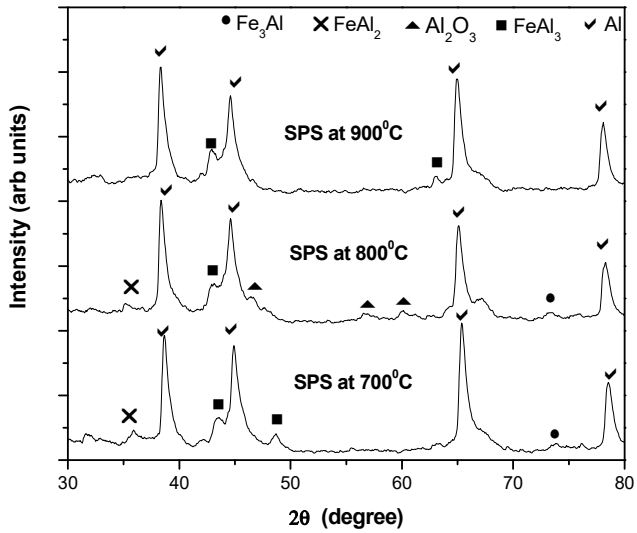

Figure 2. XRD patterns of 10 vol. % (Al_2O_3 + Fe –aluminide) reinforced Al matrix composite prepared by SPS at different temperature

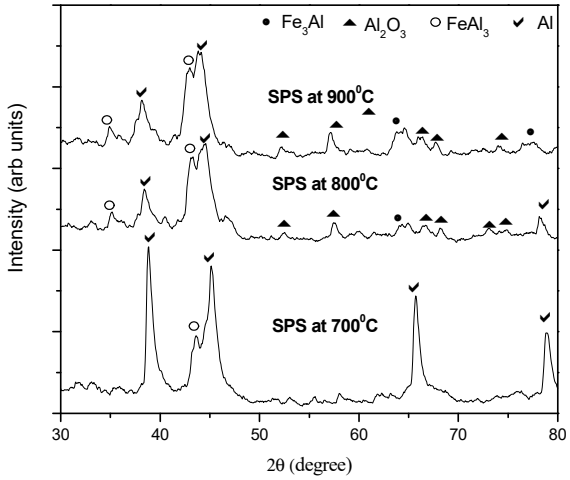

Figure 3 XRD patterns of 30 vol. % (Al_2O_3 + Fe –aluminide) reinforced Al matrix composite prepared by SPS at different temperature

The type of reinforcements formed in the composite samples with respect to the hot pressing temperature and composition were investigated using the XRD technique and the representative patterns are shown in Fig 2 and 3. XRD pattern analysis reveals the presence of $FeAl_2$, $FeAl_3$, Fe_3Al, Al_2O_3 and unreacted Al in the composite samples. The presence of the above mentioned Fe-aluminides and Al_2O_3 in the composite samples confirms the occurrence of the following in-situ reactions during SPS.

$$Fe_2O_3 + 2Al = Al_2O_3 + 2\,Fe$$

$$x\,Fe + y\,Al = Fe_xAl_y \quad (x, y \text{ can be } 1, 2 \text{ or } 3)$$

Here, it is to be noted that the initiation temperature of the above reactions with the conventional micron sized Fe_2O_3 powder is much higher compared to the hot pressing temperatures of this investigation under consideration. Therefore, we conclude that the onset temperature of the in-situ reactions has decreased significantly with the use of nanosized Fe_2O_3 powder.

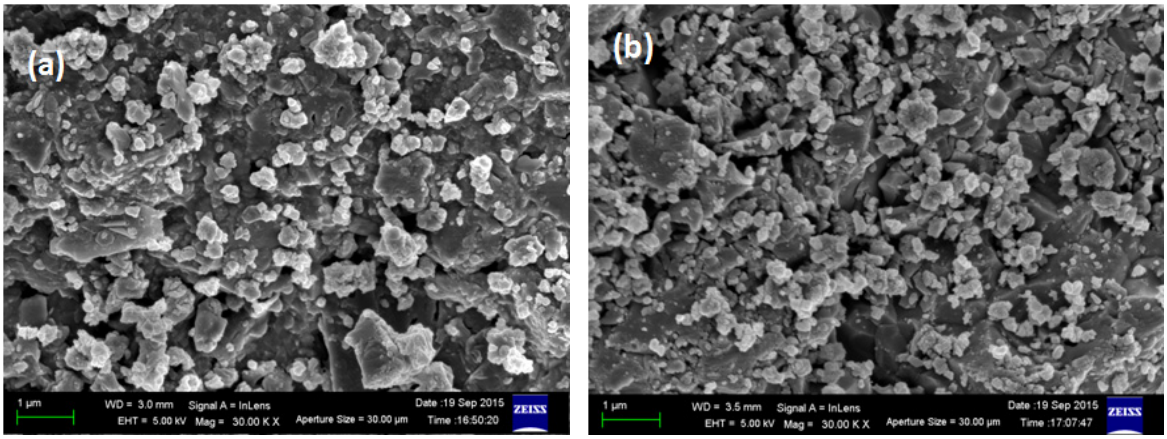

Figure 4. FESEM micrographs of (a) 10 vol.% and (b) 30 vol.% reinforced composite prepared by SPS at 900^{0}C

Representative FESEM micrographs of the in situ composite with varying reinforcement content prepared by SPS at 900^{o}C are illustrated in Figs. 4 a-b. The microstructural feature of the composites reveals the presence of very fine (less than 1 μm) and well distributed reinforcements (bright contrast) in the host aluminum matrix. EDS investigations and analysis reveal existence of Fe-aluminide and Al_2O_3 in the bright areas and aluminium in the dark areas. The size of the particulate reinforcements in the host matrix has been found to increase with the increase in reinforcement content. This has been attributed to the clustering and agglomeration effects during processing.

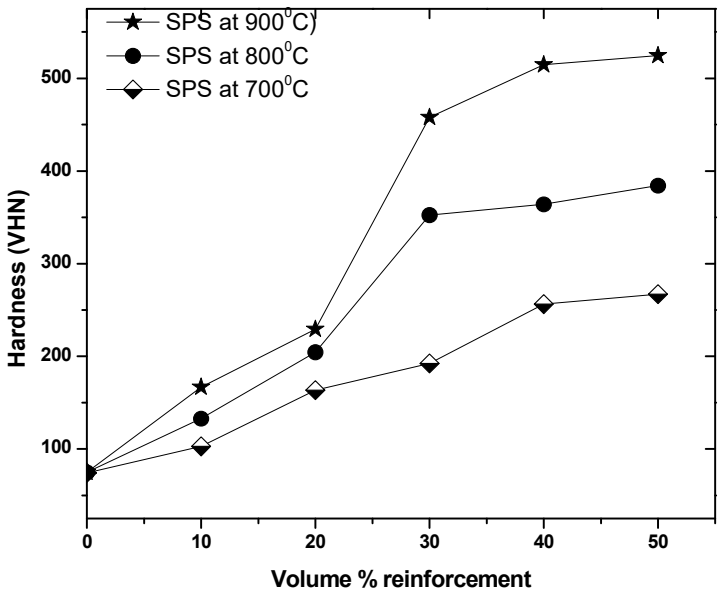

Figure 5. Hardness variation with respect to the vol. % of reinforcement phase for the composite samples prepared by SPS at 700, 800 and 900°C

Variations in the Vickers hardness values recorded on the bright and dark shaded microstructural areas as a function of temperature and reinforcement content is shown in Figure 5. The standard deviation of the hardness values recorded on the bright and dark areas has been found to be ± 10 and ± 5 VHN respectively. It has been found that the hardness increase remarkably with the increase in temperature and reinforcement content. Considering the microstructural features of the composite samples, EDS results and hardness values, we infer that the formation of hard Al_2O_3 and intermetallic phases via the in- situ reaction became kinetically more favorable with the increase in processing temperature.

4. Conclusions

Based on the investigation the main results are summarized as follows:

a) Fe-alumide and Al_2O_3 dispersed in-situ Aluminium base metal matrix nano composites can be prepared by Spark plasma Sintering of the green compacts containing nanosized Fe_2O_3 powder and commercially available aluminium powder.

b) The reinforcements are formed by the in situ exothermal reaction between nano-sized Fe_2O_3 and aluminium and during sintering.

c) The beginning temperature of the in-situ reaction decreases considerably with the use of nano-sized Fe_2O_3 powder.

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