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Effect of Ni addition on the preparation of Al_2O_3–TiB_2 composites using high-energy ball milling

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ABSTRACT

Al_2O_3–TiB_2 composites were synthesized using high-energy ball milling from starting powders containing Al, TiO_2, and B_2O_3. To explore the effect of the addition of another ductile metallic phase during milling, 15 wt.% Ni was added to a sample of the starting powders. The phase transformations and microstructure of the milled powder mixtures were investigated using X-ray diffraction and electron microscopy. The results showed that the Ni addition facilitated the mechanochemical reaction between the Al, TiO_2, and B_2O_3. Before the appearance of the Al_2O_3–TiB_2 composite, the intermediate product NiAl was formed by a gradual exothermic reaction. With continued milling, the final phases of Al_2O_3–TiB_2 and Ni were obtained.

1. Introduction

In recent years, a great deal of effort has been devoted to the development of ceramic–matrix composites. In particular, alumina, a ceramic material commonly used in high-temperature structural and wear resistant components, has been subjected to intense scrutiny. Much of this research focuses on incorporating one or more reinforcing phase(s) into an alumina matrix because the applications for monolithic alumina are limited by its low fracture toughness, brittle fracture behavior, and poor sinterability [1]. Various types of relatively soft metallic particles such as Ni, Fe, Al, Cu, Mo, and Nb have been added to alumina to improve its ductility [2–7]; on the other hand, alumina-based ceramic composites can be reinforced with hard secondary phases such as carbides, borides, and nitrides to produce novel ceramic materials with high strength and toughness for use in cutting tools [1,8–11].

Titanium diboride is a potential candidate for a secondary reinforcing phase because of its high melting point, low specific weight, high hardness, high strength to density ratio, good wear resistance, and excellent thermal and chemical stability up to 1973 K [12]. It also has good structural and thermodynamic compatibility with alumina. In a previous study, Sharifi et al. [13] synthesized Al_2O_3–TiB_2 nanocomposites from Al, B_2O_3 and Ti using a mechanochemical synthesis technique. To lower the cost of the raw materials, Sharifi et al. [14] and Khaghani-Dehahani et al. [15] developed techniques to fabricate Al_2O_3–TiB_2 composites using mechanical alloying (MA) from Al, TiO_2, B_2O_3 and Al, TiO_2, H_3BO_3, respectively. The aluminothermic reduction reaction accelerated the formation of the Al_2O_3–TiB_2 composite, and in the final product the two phases were homogeneously distributed. Taken together, these studies conclusively demonstrated that MA is an effective method to synthesize Al_2O_3–TiB_2 ceramic composites.

MA is a solid-state powder processing technique involving the repeated welding, fracturing, and re-welding of powder particles in a high-energy ball mill. Lü et al. [16] proposed that the raw materials used in MA should include at least one ductile metal to act as a host or binder to hold together the other ingredients. The powders deform plastically under high-energy collision. With new clean surfaces exposed by the mechanical treatment, the powders weld to each other to form new, aggregate particles with different compositions. These welding and fracturing processes are repeated until a fully alloyed powder is formed. Efficient cold-welding of the powders is essential to ensure complete mechanical alloying.

In view of these successful applications of MA to the preparation of Al_2O_3–TiB_2 composites and the determination of the essential conditions required, the authors have ever added the ductile metal Ni to the raw Al, TiO_2, and B_2O_3 powders to prepare the Al_2O_3–TiB_2 composite powders [17]. Based on the study, in this paper, a control group without Ni was milled under the same condition and the results showed that the Ni addition facilitated

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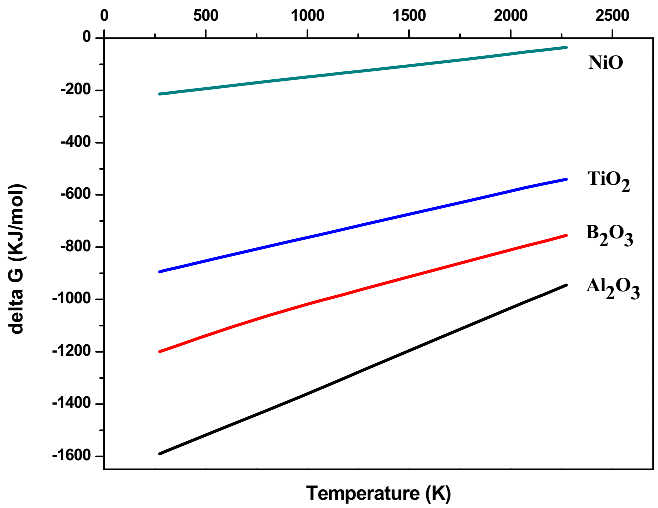

Fig. 1. Standard Gibbs free energy of formation as a function of temperature for Al_2O_3, B_2O_3, TiO_2 and NiO.

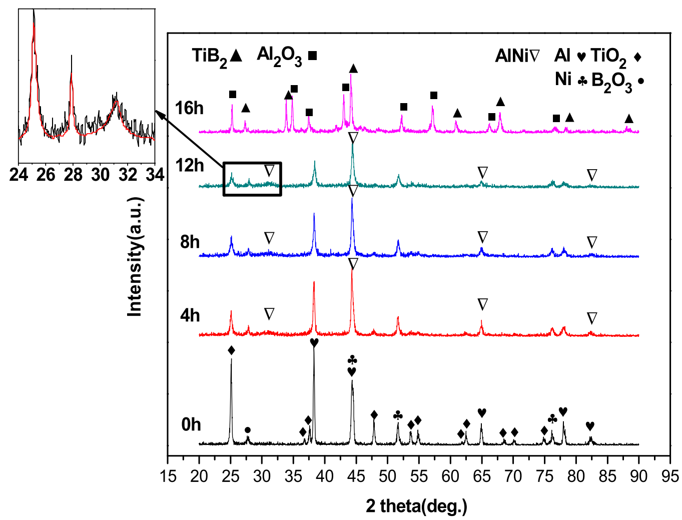

Fig. 2. XRD patterns of the Ni-containing powders after different milling times. The inset picture shows corresponding local amplification.

the mechanochemical reaction between the Al, TiO_2, and B_2O_3. Additionally, the formation mechanism of Al_2O_3–TiB_2 composite and the effect of Ni on the MA process were investigated.

2. Experimental procedure

Al (99% purity), TiO_2 (99% purity), B_2O_3 (99% purity), and Ni (99% purity) powders were used as raw materials. The mixture of Al, TiO_2, and B_2O_3 accounted for 85% of the total powder weight, and was prepared in accordance with the stoichiometry of the reaction:

$$10Al + 3TiO_2 + 3Br_2O_3 = 5Al_2O_3\,(71\,wt.\%) + 3TiB_2\,(29\,wt.\%),$$

$$\Delta G^{\circ}_{298} = -2633.9\,kJ/mol, \quad \Delta H^{\circ}_{298} = -2710\,kJ/mol \qquad (1)$$

An addition of 15 wt.% Ni was added to this mixture to make the "Group #1" powders. For comparison purposes, raw powders without any Ni addition were also made ("Group #2").

Ball milling of the powder mixtures was carried out in a planetary ball mill with a stainless steel vial (150 mL) and balls at room temperature under an Ar atmosphere. Since the highest collision energy can be obtained if balls with different diameters are used [18], two different sizes of balls (⌀8 mm and ⌀6 mm) were employed in a weight ratio of 1:1. The ball-to-powder weight ratio and the rotational speed of the vial were 10:1 and 500 rpm, respectively. The milling was interrupted after predetermined intervals, and small amounts of powder were removed for characterization.

Phase transformations in the powder mixture were analyzed using X-ray diffractometry (XRD). The morphology and microstructure of the milled powder particles were examined using transmission electron microscopy (TEM). The TEM sample was prepared by suspending the powder sample in ethanol and subjecting it to ultrasonic vibration. A drop of the suspension was then placed on a carbon-coated copper grid and dried.

3. Results and discussion

The Ni-free Al_2O_3–TiB_2 nanocomposites were synthesized according to Reaction (1) by high-energy ball milling with a self-propagating combustion mode [14]. For the specimen with the 15 wt.% Ni addition, the reaction became more complex. Since the Gibbs free energy of NiO is higher than that of both B_2O_3 and TiO_2 (Fig. 1), neither the Ni/B_2O_3 nor the Ni/TiO_2 reactions occur.

Table 1
Lattice parameter (a) of NiAl for different milling times.

Milling time (h)	4	8	12
a (Å)	2.877	2.871	2.867

This suggests that Ni would only react with Al to form an Al–Ni intermetallic compound before aluminothermic Reaction (1) took place during the mechanical milling. In addition, studies on the preparation of Al–Ni alloys by MA of elemental powders had been reported [19–21]. Mashreghi and Moshksar [20] reported that a NiAl intermetallic compound was formed during MA of $Al_{50}Ni_{50}$ according to the gradual exothermic reaction:

$$Al + Ni = NiAl, \qquad \Delta G^{\circ}_{298} = -117.2\,kJ/mol,$$

$$\Delta H^{\circ}_{298} = -118.4\,kJ/mol \qquad (2)$$

Fig. 2 shows the XRD patterns of the samples with the 15 wt.% Ni addition after milling for 0, 4, 8, 12, and 16 h. The diffraction pattern of the un-milled powder shows peaks associated with the raw materials: Al, Ni, TiO_2, and B_2O_3. B_2O_3 was represented in only one sharp peak at around $2\theta = 28^{\circ}$ due to the overall lower intensity of B_2O_3 peaks compared to other components. After 4 h of milling, the peaks corresponding to the raw powders decreased in intensity and broadened; this is caused by the reduction of the crystallite size and the microstrain induced in the powders. Simultaneously, a small amount of the NiAl phase was detected. As the amount of Ni added into the raw mixture was small and only a small fraction of the Ni participated in the gradual exothermic Reaction (2), the peak related to NiAl phase was not remarkable. With continued milling to 12 h, the lattice parameter of NiAl decreased from 2.877 to 2.867 Å. Table 1 shows the variation in the lattice parameter of the NiAl phase during mechanical alloying. It is believed that, due to the high Al content of the raw powders, Al dissolves in the NiAl phase during MA so that the lattice parameter decreases [21].

Peaks associated with the Al_2O_3 and TiB_2 phases were detected after 16 h of milling. Meanwhile, the peak associated with the raw materials (Al, TiO_2, B_2O_3, Ni) and the intermediate product (NiAl) disappeared almost entirely. This suggested that, in spite of the presence of the dilutant Ni, Reaction (1) still took place with a combustion mode. Additionally, the intermediate product NiAl