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Microstructure and mechanical properties of Al_2O_3–MgB_2 composites

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Abstract

The microstructure and mechanical properties of Al_2O_3–5wt.% MgB_2 composites were studied by scanning electron microscopy (SEM) and Vickers hardness measurements. The experiments show that MgO whiskers can grow at temperatures as low as 950 °C. When sintered at 1450 °C, due to the strong vaporization of Mg, MgO and $MgAl_2O_4$ appear around Al_2O_3 grains. The microstructure of the Al_2O_3–5wt.% MgB_2 differs remarkably from Al_2O_3 sintered at the same temperature. The fracture toughness of the Al_2O_3–5% MgB_2 composite is 4.0 MPa m$^{1/2}$ which is slightly higher than for pure Al_2O_3 ceramics.

Keywords: B. Microstructure; D. Alumina; MgB_2

1. Introduction

Alumina is a widely used ceramic due to its refractoriness, high hardness, good wear resistance and high chemical stability, however, the brittleness limits its applications. It has been shown that the introduction of a second phase can reduce Al_2O_3 grain size and enhance fracture toughness (K_{1C}) significantly [1–4]. MgO is a traditional additive to Al_2O_3 since it can reduce the sintering temperature and grain size. In early studies on Al_2O_3–MgO composites, MgO was added directly to Al_2O_3. However, the enhancement of the K_{1C} is not obvious. Recently, it has been reported that nanometer-sized MgO whiskers can grow during the oxidation of MgB_2 at 900 °C [5,6]. Since the MgO whisker has high melting point, high strength and good plasticity, adding MgB_2 may be a desirable way to grow MgO whiskers in Al_2O_3 and enhance its toughness. In this work, we investigate an Al_2O_3–5% MgB_2 composite and discuss the influence of adding MgB_2 on the microstructure and mechanical properties.

2. Experimental

MgB_2 was synthesized by vacuum encapsulation. A stoichiometric mixture of commercial Mg (purity > 99%, 100–200 mesh) and B powder (purity > 96%, average particle size ∼2 μm) was ground in an agate mortar, then pressed into pellets and put into a quartz tube which was vacuum encapsulated. The sample was heated rapidly to 850 °C and held at this temperature for 2 h. The obtained MgB_2 sample was checked by X-ray diffraction (XRD) and scanning electron microscopy (SEM). MgB_2 powder was heated in a thermal-analyzer to 900 °C and kept at this temperature for 3 h, then examined by SEM.

MgB_2 powder was added into Al_2O_3 powder (purity > 99%, average particle size 1.4 μm) at a weight ratio 5:95, and then mixed thoroughly and pressed into pellets under 10 mPa. The pellets were put into the MgO crucible and sintered at a heating rate of 5–10 °C min^{-1} in an electric furnace and kept at the final temperature (850, 950 and 1450 °C, respectively) for 1 h. Pellets of pure Al_2O_3 and Al_2O_3–5%MgO were also sintered at the same conditions in order to comparing their microstructures with Al_2O_3–5% MgB_2 composite.

The microstructure was studied with a SEM equipped with X-ray energy dispersive analysis (EDS). The surface

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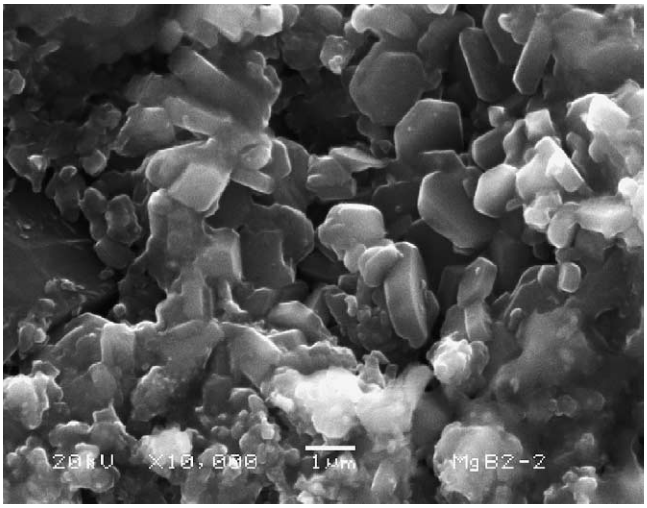

Fig. 1. Microstructure of MgB_2 made by vacuum encapsulation.

of the pellets was burnished by SiC sand paper, and then polished by diamond cream to get smoothness. Vickers diamond hardness indentation at a load of 9.8 N and loading time of 10 s was used to test the mechanical properties, and fracture toughness was calculated by using the average length of radial cracks and the Liang's equation [7].

3. Results and discussion

Fig. 1 shows the microstructure of the synthesized MgB_2. It can be seen that many grains show hexagonal shape due to the hexagonal crystal structure of MgB_2. XRD pattern shows that the sample prepared by vacuum encapsulation has a pure phase of MgB_2 with a thin impurity of MgO that may be caused by the thin residual oxygen left in the quartz tube. After heating at 900 °C for 3 h in air, MgO whiskers with diameters about 10–100 nm appear on the grain surface in the oxidation process of MgB_2, as Fig. 2 shows. The

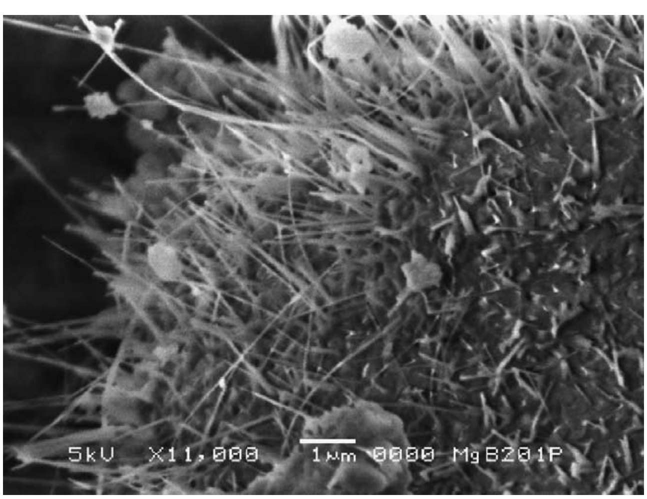

Fig. 2. MgO whiskers growing from oxidation of pure MgB_2.

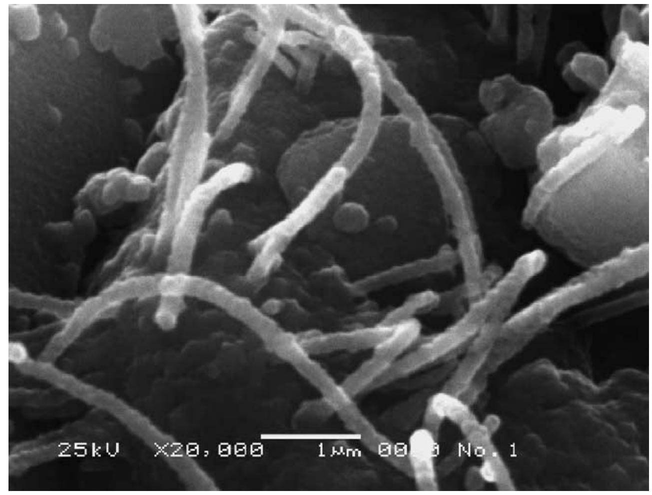

Fig. 3. Micrograph of the Al_2O_3–5% MgB_2 composites sintered at 850 °C.

growth of the whisker is caused by the reaction of oxygen with the vapor of Mg [8]. When MgB_2 is heated in air or in oxygen, MgO can be formed by two ways. In the first way, oxygen diffuses into the grain of MgB_2 and reacts with Mg to form MgO. In the second way, Mg evaporates first and then reacts with oxygen on the grain surface to form MgO. The already formed MgO on the grain surface will serve as a seed crystal (as can be seen from Fig. 2, there are many small prominences on the grain surface) and then MgO will grow continuously on the seed to form long thin MgO whiskers.

When MgB_2 is added into Al_2O_3 and sintered at 850, 950 and 1450 °C, its microstructure is remarkably different from that of the pure MgB_2 after oxidation. Fig. 3 shows the micrograph of Al_2O_3–5% MgB_2 sintered at 850 °C. Comparing with Fig. 2, we can see that the diameter of the whisker becomes thicker and almost does not vary when the whisker grows longer. It can also be seen that all whiskers in Al_2O_3–5%MgB_2 sintered at 850 °C have approximately the same diameter of about 200 nm. On the other hand, whiskers grown from pure MgB_2 are relatively thin. In addition, from Fig. 2, we can find that some whiskers become thinner gradually when they grow longer. The other obvious difference is that the whiskers grown in Al_2O_3–5% MgB_2 composite have bamboo shape and the number of whiskers in Al_2O_3–5% MgB_2 composite is less than for the pure MgB_2.

It is well-known that most of the whiskers can only grow in the vapor phase. Therefore, growth environment, such as temperature, time, atmosphere, and impurity concentration have very important effects on the diameter, the length and the shape of the whiskers. In the above two experiments of growing MgO whiskers from MgB_2, temperature and heating time are similar, so the difference of the whisker shape is caused by the difference of oxygen atmosphere and impurities. When MgB_2 is put into Al_2O_3 powder and pressed into pellets, it is difficult for the oxygen to enter the sample. This will result in oxygen insufficiency in the sample that depresses the oxidation process of MgB_2 and affects

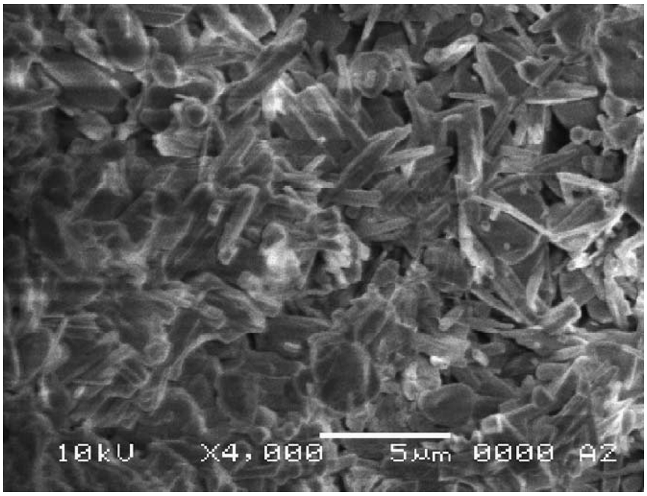

Fig. 4. Micrograph of the Al_2O_3–5% MgB_2 composites sintered at 950 °C.

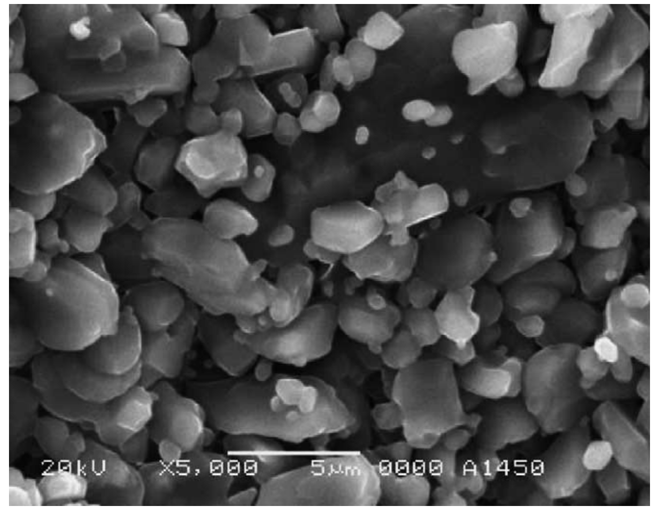

Fig. 6. Micrograph of the alumina without MgB_2 additive sintered at 1450 °C.

the MgO whisker shape. In the case of oxygen deficiency, in MgO crystal seed formed on the grain surface dislocations may be generated, and the bamboo node of the whisker may be the boundary of a twin-crystal. If sintering temperature increases to 950 °C, vaporization and oxidation of Mg in MgB_2 will become very strong. The bamboo node shape MgO whiskers grow up and become long thin rods with sub-micron size as shown in Fig. 4.

When Al_2O_3–MgB_2 samples are sintered at 1450 °C, their microstructure changes remarkably from samples sintered at 850 and 950 °C. Fig. 5 shows the micrograph of Al_2O_3–5% MgB_2 heated at 1450 °C, which indicates that most of the grains in the sample grow into columnar shape grain. The oxidation of MgB_2 plays an important role in the formation of columnar shape grains. At high temperature, Mg elements vaporize almost and Mg vapor can oxidize to form MgO and $MgAl_2O_4$ around Al_2O_3 grains. This causes formation of columnar shape grains. Without MgB_2 additive the columnar shape grain cannot be formed. Fig. 6 shows the micrograph of monolithic Al_2O_3 ceramic without MgB_2 additive; it can clearly be seen that there are no columnar shape grains. Therefore, it is the addition of MgB_2 that helps the formation of columnar shape grains in the Al_2O_3–5% MgB_2 composites.

It is well-known that the addition of MgO can promote liquid phase sintering of Al_2O_3 ceramics, thus resulting in a uniform microstructure and reducing the anisotropy of alumina [9]. However, the effect of adding MgB_2 into Al_2O_3 is not as the same as adding MgO directly. Fig. 7 shows the micrograph of Al_2O_3 with 5% MgO additive. The liquid phase sintering effect is obvious, but the columnar shape grains are rare comparing with the samples with MgB_2 additive. The main difference between MgB_2 and MgO additive may be explained as follows. When adding MgO directly, only liquid phase sintering effect exists and the vaporization of Mg is difficult due to the high stability of MgO at high

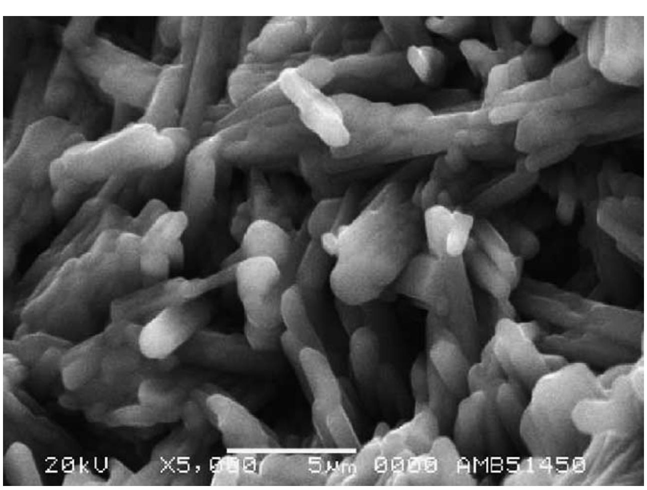

Fig. 5. Micrograph of the Al_2O_3–5% MgB_2 composites sintered at 1450 °C.

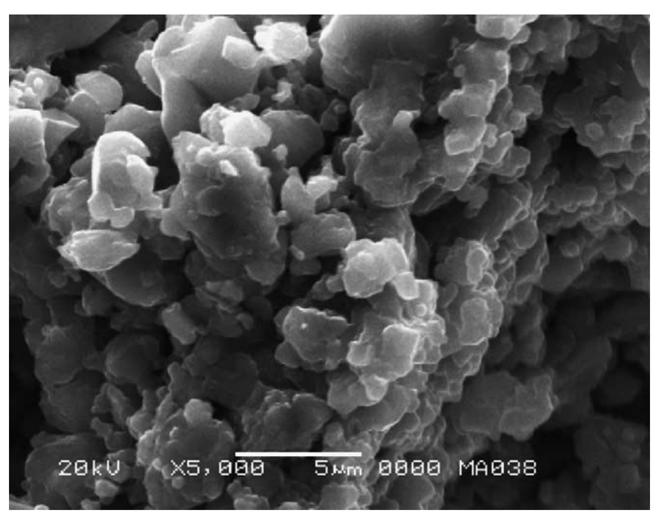

Fig. 7. Micrograph of the alumina with 5% MgO additive sintered at 1450 °C.

Table 1
Mechanical properties of Al_2O_3 and Al_2O_3–5% MgB_2 ceramics

Specimen	Density ($g\,cm^{-3}$)	Hardness (H_v) GPa	Fracture toughness (K_{1C}) $mPa\,m^{1/2}$
Al2O3–5%MgB2	3.58	2.09	4.0
Al_2O_3	3.62	2.26	3.3

temperature. While in the case of adding MgB_2, Mg can vaporize easily and form a uniform distribution in the specimen. When Mg vapor meets oxygen, MgO and/or $MgAl_2O_4$ will grow on the boundary of Al_2O_3 grain and a certain direction may be preferred in the continuous growth, thus columnar shape grains are formed. It was pointed out [10,11] in pure Al_2O_3 abnormal growth of grain is not observed. However, with the help of some additive (such as Fe_2O_3, TiO_2, and MnO_2 [12]) some liquid phase can be formed resulting in abnormal growth of grains, i.e., in some directions the growth rate of grains is faster than other directions. Our experiment shows that MgB_2 is a much effective additive that helps abnormal growth and formation of columnar shape grains.

It is believed that the formation of columnar shape grains can increase the fracture toughness. Table 1 compares some mechanical properties of Al_2O_3–5% MgB_2 and Al_2O_3 composites, and shows that their hardness and density are similar, but the fracture toughness of Al_2O_3–5% MgB_2 composite increases slightly.

The increase of fracture toughness of the Al_2O_3–5% MgB_2 composite is mainly due to two factors, i.e., (i) liquid phase sintering and (ii) the formation of columnar shape grains. At high temperature, MgB_2 is oxidized strongly and can form a uniform Mg vapor around Al_2O_3 grains, which is a much effective way of liquid phase sintering and is better than adding MgO directly. The formation of columnar shape grains can also enhance fracture toughness because columnar shape grains can form bridge that increases the fracture resistance.

4. Conclusions

By controlling the environmental condition, such as temperature and oxygen atmosphere, MgO whiskers with different shape can be obtained by adding MgB_2 to Al_2O_3. When the sample is sintered to 1450 °C, due to the strong vaporization of Mg, MgO and/or $MgAl_2O_4$ will grow around Al_2O_3 grains giving rise to columnar shape grains. The fracture toughness of the Al_2O_3–MgB_2 composites is higher than that of pure Al_2O_3. The advantage of the MgB_2 additive compared to MgO is that the MgB_2 can form a uniform Mg vapor, help the formation of columnar shape grains and increase the fracture toughness of Al_2O_3–MgB_2 composite.

Acknowledgements

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