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Preparation and Electrical Properties of MWNTs/Al_2O_3 Ceramics Composites†

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In this work, MWNTs/Al_2O_3 composites were prepared by mixing multi-walled carbon nanotubes (MWNTs) and alumina. The MWNTs/Al_2O_3 composites were obtained by hot pressing sintering in vacuum atmosphere, which had good electric conductivity, the addition of carbon nanotubes were 0.1 wt % alumina, 0.3 wt % and 0.5 wt %. The results indicated that the MWNTs/Al_2O_3 composites, which were formed by addition of 0.3 wt % carbon nanotubes, had the minimum resistivity value and the resistivity value could reach to 101 Ω cm. Compared with the pure Al_2O_3, the resistivity value lower 5 times. The SEM pictures showed that carbon nanotubes wrapped alumina particles in the form of aggregates. They formed a conductive network in the alumina grain boundaries, so that they could lower resistivity value and improve its electrical conductivity. We also analyzed the MWNTs/Al_2O_3 composites conductive mechanism and showed the conductivity model.

Key Words: MWNTs/Al_2O_3, Conductive property, Composites, Resistivity.

INTRODUCTION

Carbon nanotubes have been a hot focus in the field of the researches of materials[1]. Carbon nanotubes have superior mechanical properties and excellent electrical properties. The elastic modulus of multi-walled carbon nanotubes can reach 1.8 T Pa and bending strength can reach 14.2 GPa. The longitudinal electrical conductivity of single-walled carbon nanotubes can reach 106 S/m at room temperature, therefore, the carbon nanotubes have great potential in composite materials field.

So far, there are many reports about carbon nanotube composites, however, most researchers focus attention on reinforced CNTs/polymer materials[2-4] and CNTs/metal materials[5-7]. The work on the enhancement of non-metallic mineral matrix is still in the exploratory phase. The follow-up results studied by Zhan *et al.*[8] showed that the electrical conductivity of SWCNT/Al_2O_3 would increase with the increasing of carbon content. The electrical conductivity of 15 vol % SWCNT/Al_2O_3 can arrive at 3345 S/m. Flahaut[9] found that ceramic and metal oxide matrix can turn into conductor by adding carbon nanotubes in and the electrical conductivity values were between 0.2 and 4.0 S/m. The conductivity value was related to the damaged degree of carbon nanotubes which were in the matrix, when the structure of nanotubes were damaged, the composites could not transmit electricity. Now, there are mainly two main problems on the research of the MWNTs/Al_2O_3 composites, first, the dispersion of carbon nanotubes in the matrix. Second, the compatibility between carbon nanotubes and the alumina matrix is poor and the interface strength between the two is also poor. In this experiment, solvent system was used to mix carbon nanotubes and alumina directly and the alumina matrix was formed by the solid phase reaction of alumina and so, the MWNTs/Al_2O_3 composites were obtained.

EXPERIMENTAL

Multi-walled carbon nanotubes were produced by Shenzhen Nano-Port Co., Ltd, the purity greater than 95 %, inside diameter is 5-20 nm and outside is 15-40 nm. Length/diameter >100, the density is *ca.* 2.0 g/cm^3. The multi-walled carbon nanotubes were prepared for the next procession,which were treated by concentrated nitric acid for 4 h at 110 ºC.

Alumina powders were the M100 which were produced by Hai Tian Feng Ceramics Factory (raw particle size 10-20 nm).

PEG2000 produced by Shanghai Chemical Reagent Company Chinese Medicine Group was used as dispersant, anhydrous ethanol (analytical reagent) was used as dispersion medium.

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Preparation of MWNTs/ Al_2O_3 powders: The addition of the quantity of carbon nanotubes were 0.1 wt % alumina, 0.3 wt %, 0.5 wt %, weighed required MWNTs to put in 1 wt % (1 wt % MWNTs) PEG solution and alumina powders put in ethanol solution, then to stir about 10 min with the electromagnetic agitator and then with ultrasonic dispersing for 10 min, so that the two dispersed suspension were formed. Alumina powder suspension were added dropwise to carbon nanotubes suspension, which were fast stirring and then filtered and dried at 100 ºC and then filter through 200 meshes after grinding. The mixed powder were hot pressed sintering in atmosphere of 1 atm flowing of pure Ar. Axial pressure was 3.2 t, so the pressure on the sample was 30 MPa. Holding time was 0.5 h.

RESULTS AND DISCUSSION

Resistance and resistivity of MWNTs/Al_2O_3 composites: The resistivity decreased rapidly with the gradual addition of carbon nanotubes (Fig. 1) indicating that the composites had conductive properties. When the content reached 0.3 wt %, the composites resistivity were minimum (101 Ω cm) and compared with pure Al_2O_3 (4.54E+07), the resistivity value lower 5 times (Table-1).

TABLE-1
RESISTANCE AND RESISTIVITY OF MWNTs/Al_2O_3 COMPOSITES

Wt/%	0 MWNTs	0.1 MWNTs	0.3 MWNTs	0.5 MWNTs
Resistance (Ω)	1.14E+09	1.21E+08	2.53E+03	8.20E+03
Resistivity (Ω cm)	4.54E+07	4.84E+06	1.01E+02	3.27E+02

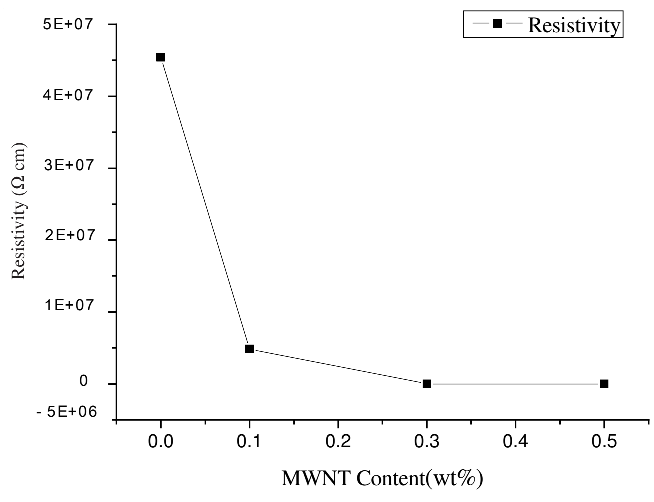
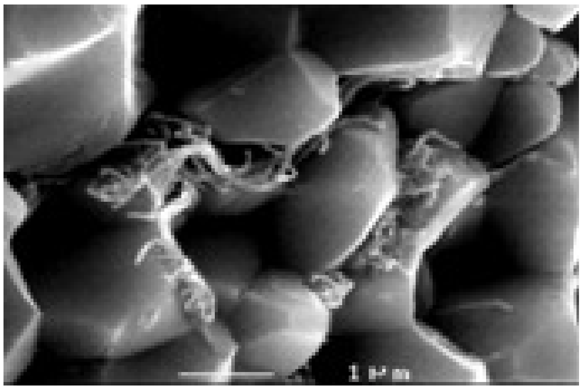
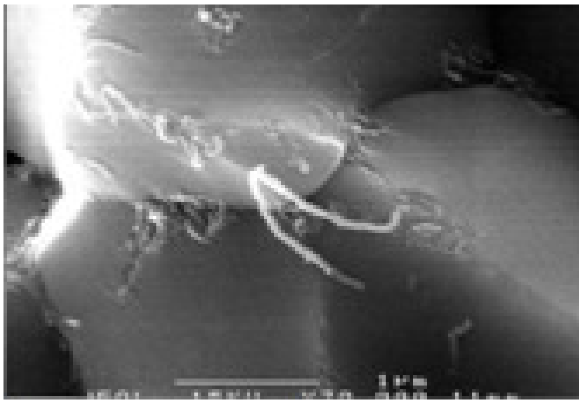


Fig. 1. Effect of multi-walled carbon nanotubes content on the resistivity of MWNTs/Al_2O_3 composites

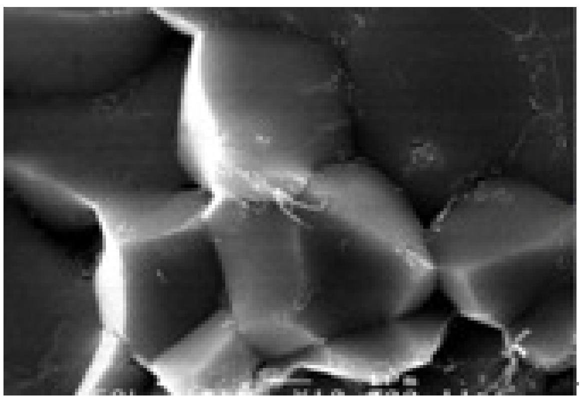
Conducting Mechanism of MWNTs/Al_2O_3 composites: From the fracture surface of MWNTs/Al_2O_3 composites, we found that 0.3 wt % MWNTs/Al_2O_3 composites could conduct electricity due to carbon nanotubes wrapped alumina particles in the form of aggregates, which formed a conductive network in the alumina grain boundaries [Fig. 2(a)]. The conductive network connected to each other in the MWNTs/Al_2O_3 composites, so it had good conductivity. From different directions of the fracture surfaces of 0.5 wt % MWNTs/Al_2O_3 composites (Fig 2(b) and (c)), we could see the details of the alumina grains coated with carbon nanotubes. And the carbon nanotubes connected to each other due to its arrangement, which was the necessary condition for transmiting electricity. So it is concluded that there were two very important conditions for good conductivity, first, as completely as alumina grain boundaries coated by carbon nanotubes to ensure continuous conduction; second, carbon nanotubes should arrange along the grain boundaries to ensure that carbon nanotubes had many chances to contact each other, which could allow current to pass from one carbon nanotube to another.



(a) 0.3WMNTs



(b) 0.5WMNTs



(c) 0.5WMNTs

Fig. 2. SEM image of the fracture surface of MWNTs/Al_2O_3 composites

Relationship between the microstructure and electrical property of MWNTs/Al_2O_3 composites: From above analyses we could see that what was the role of the carbon nanotubes played had much to do with its shape in MWNTs/Al_2O_3 composites. If the carbon nanotubes within the alumina grains and embedded within the two grains, the mechanical properties of carbon nanotubes could perform excellently, which could improve the flexural strength. If carbon nanotubes could arrange along the grain boundaries to ensure that carbon nanotubes had many chances to contact each other, which could allow current to pass from one carbon nanotube to another, the MWNTs/Al_2O_3 composites would have better electric conductivity. The relationship between the microstructure and electrical property of MWNTs/Al_2O_3 composites could be depicted by two ideal models (Fig. 3).

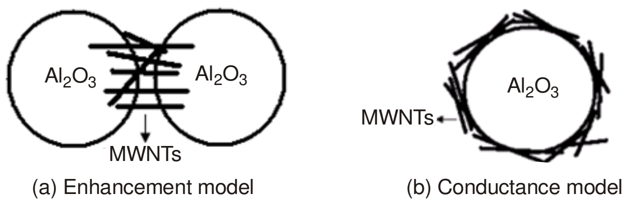


Fig. 3. Model of enhancement and conductance of MWNTs/Al_2O_3 composites

In addition, from the view of the fracture surface of MWNTs/Al_2O_3 composites, we could see that carbon nanotubes reunited together and most of the carbon nanotubes had been cut short. The ratio of height to diameter of carbon nanotubes was not well used which was realated to the method of the preparation of the MWNTs/Al_2O_3 composites. Firstly, the purified time of carbon nanotubes may be too long and so the carbon nanotubes were cut too short, which caused the ratio of height to diameter of carbon nanotubes to useless. Secondly, because the Al_2O_3 were mixed with carbon nanotubes in the form of particles and could find that the largest particle reached *ca.* 25 μm.The carbon nanotubes maintained their shapes in the sintering process and did not like metal particles which can diffuse, grow up, transfer and hinder the growth of grains. In addition, the carbon nanotube reunions were opened by ultrasonic dispersion temporarily, but carbon nanotubes would subside and lap during the process of dryness, Therefore, the carbon nanotubes would not disperse well in the alumina matrix (Fig. 4).

So that, although with the addition of carbon nanotubes in the MWNTs/Al_2O_3 composites, the bending strength and electrical properties were improved, however, if the carbon nanotubes could disperse in the alumina matrix well, the bending strength and electrical properties of the MWNTs/Al_2O_3 composites should be great improved.

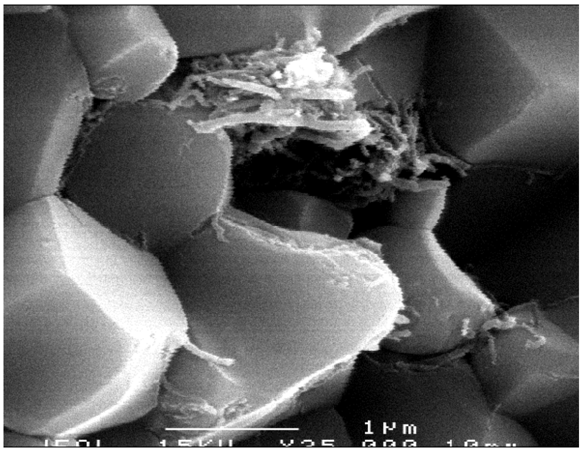


Fig. 4. SEM image of the fracture surface of 0.3 wt % MWNTs/Al_2O_3 composites

Conclusion

Alumina matrix grains would refine and grow uniformly with the addition of carbon nanotubes. The MWNTs/Al_2O_3 composites were obtained by hot pressing sintering in vacuum atmosphere, which would have good electric conductivity. The results indicated that the MWNTs/Al_2O_3 composites, which were added 0.3 wt % carbon nanotubes, had the minimum resistivity value, and the resistivity value can reach to 101 Ω cm. Compared with the pure Al_2O_3, the resistivity value lower 5 times. The SEM pictures showed that carbon nanotubes wrapped alumina particles in the form of aggregates, they formed a conductive network in the alumina grain boundaries, so it can lower resistivity value and improve its electrical conductivity.

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