Preparation and Properties of Epoxy Resin Based Conductive Composites

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Abstract: The epoxy resin-based conductive composite was prepared by filling the epoxy resin with carbon fiber powder and graphene. The influence of the additive quantity on the conductive properties and mechanical properties were investigated, and the influence of carbon fiber powder surface treatment on the conductive properties and mechanical properties were investigated. The results show that the addition of carbon fiber powder and graphene can improve the conductivity and mechanical properties of epoxy composites.

1. Introduction
Epoxy resin (EP) is an insoluble thermosetting plastic. The cured EP has excellent physical and chemical properties. It has many advantages, such as high adhesion, small volume shrinkage, good electrical insulation, high mechanical strength, easy processing and so on[1]. With these advantages, EP occupies a large part of the thermosetting market. EP itself is non-conductive. In recent years, it has become a hot topic to add conductive filler to prepare conductive composites. At present, the widely used and promising conductive composites are the preparation of carbon based fillers, which mainly include carbon fiber, graphene, carbon nanotubes and so on[2-3]. With the development of carbon fiber, the research and development of carbon fiber conductive composites have been widely concerned in the world. In this paper, carbon fiber powder, graphene and EP were used to prepare conductive composites by hot pressing. The effects of filler content and surface treatment of carbon fiber powder on the electrical and mechanical properties of the composites were studied.

2. Experimental part

2.1 Experimental drugs
EP, Nantong Xingchen synthetic material limited company; Carbon fiber powder, 400 mesh, Yancheng Xiangsheng Carbon Fiber Technology limited company; Graphene, Nanometer scale, Yantai neneng New Material limited company; Methylhexahydrophthalic anhydride; Shanghai McLin Biochemical Technology limited company; 2,4,6-tris (Dimethylaminomethyl) phenol, Kunshan jiulimei Electronic Materials limited company; Silane coupling agent (KH-570), Jinan xingfeilong Chemical limited company.

2.2 Experimental instruments
Vacuum drying oven, DHG-9140A, Shanghai Yiheng Scientific Instrument Co., Ltd; Planetary mixer, SR-2000, Dongguan ruisimai Electromechanical Technology Co., Ltd; Hand held DC low resistance tester, AT518, Changzhou anbai Precision Instrument Co., Ltd; Digital cantilever impact tester, XJU, Xiamen Chongda Intelligent Technology Co., Ltd; Fourier transform infrared spectrometer, Nicolet-6700, Nicolet company; Thermogravimetric analyzer, Q50, TA instrument.
2.3 Sample preparation
Weigh a certain amount of EP and conductive filler. Heat EP grease to 60℃, add conductive filler and stir for 20 minutes, then add curing agent and accelerator and stir for 20 minutes. After mixing, vacuum filter at 80℃ for 25 minutes, take it out, pour it into the preheated mould, and put it into the oven for curing at 120℃ for 2 hours.

2.4 Testing and characterization
The volume resistivity was measured by a hand-held DC low resistance tester with a sample size of 20 mm×30mm×5mm. The impact performance was tested by cantilever impact testing machine, and the sample size was 10 mm×50mm×5mm, notch 2mm. The thermal stability was tested by Q50 thermogravimetric analysis of TA company, the sample was 5-10mg powder. The powder before and after surface treatment was analyzed by Nicolet 6700 Fourier transform infrared spectrometer.

3. Results and discussion

3.1 Effect of graphene addition on electrical conductivity of composites
The electrical conductivity of the composite is shown in Figure 1.

![Figure 1](image1)

Figure 1 Volume conductivity of composites with different graphene content

It can be seen from Figure 1 that the conductivity of pure EP is 0 S/m, which is still insulating. When the content of graphene is about 0-5wt%, the composite is still insulating. When the addition of graphene continues to increase to 10wt%, a local electric network is formed, and the evacuation state of the system is not very complete, and the conductivity increases to 1.396×10⁻⁵S/m. When the addition amount is 15wt%, the maximum conductivity of the composite can reach 2.543×10⁻⁴S/m. At this time, the dispersion state of graphene is improved, so the conductivity is greatly improved.

3.2 Effect of graphene addition on impact strength of composites
The impact strength of the composites prepared by adding different contents of graphene and epoxy resin is shown in Figure 2.
According to figure 2, the impact strength of pure EP is 1.87 kJ/m², and the material shows brittleness. The impact strength of the composites with graphene is 17-24% higher than that of pure epoxy resin composites. These test data show that the toughness of epoxy resin can be improved by adding graphene in a certain range. The C = C structure of epoxy resin endows it with rigidity. In order to improve the mechanical properties of materials, increasing the crosslinking density is an effective way. High crosslinking density can reduce the fracture resistance. Graphene nanoparticles can improve the toughness of the composites with appropriate addition.

3.3 Effect of graphene and carbon fiber powder on properties of composites

3.3.1 Effect of carbon fiber powder addition on electrical conductivity of composites

In this experiment, when the content of graphene is set, the conductivity does not change obviously when the carbon fiber powder is added below 5wt%. When the carbon fiber powder is added to 15wt%, the conductive path is the best, and the conductivity reaches $1.296 \times 10^{-4}$ S/m, which is an order of magnitude of the conductivity when adding 15wt% graphene. With the addition of carbon fiber powder, the conductivity decreased sharply when the amount of carbon fiber powder was 20wt%. The reason is that it is difficult to disperse when two kinds of powder fillers are added at the same time. When the filling amount reaches a certain critical value, agglomeration will occur and the resistance to current in the volume will increase. The composite filled with spherical powder has higher percolation threshold and lower critical index, and the possibility of agglomeration of large size filler is smaller than that of small size filler[3].

3.3.2 Effect of carbon fiber powder addition on impact strength of composites

In this experiment, when the content of graphene is set, the conductivity does not change obviously when the carbon fiber powder is added below 5wt%. When the carbon fiber powder is added to 15wt%, the conductive path is the best, and the conductivity reaches $1.296 \times 10^{-4}$ S/m, which is an order of magnitude of the conductivity when adding 15wt% graphene. With the addition of carbon fiber powder, the conductivity decreased sharply when the amount of carbon fiber powder was 20wt%. The reason is that it is difficult to disperse when two kinds of powder fillers are added at the same time. When the filling amount reaches a certain critical value, agglomeration will occur and the resistance to current in the volume will increase. The composite filled with spherical powder has higher percolation threshold and lower critical index, and the possibility of agglomeration of large size filler is smaller than that of small size filler[3].

The change of impact strength with the addition of carbon fiber powder is shown in Figure 4.
As shown in Figure 4, it can be seen that with the addition of carbon fiber powder, the impact strength of the material increases between 0-15wt%, and reaches the best value at 15wt%, and the impact strength can reach 1.42kJ/m², which is 7.5% higher than that without carbon fiber powder. Then the impact strength decreased with the addition of carbon fiber powder.

3.4 Effect of surface treatment of carbon fiber powder on properties of composites

3.4.1 Effect of surface treatment of carbon fiber powder on electrical conductivity of composites
The surface treatment of carbon fiber powder with silane coupling agent KH-570 shows the influence on the conductivity of composite as shown in Figure 5.

Figure 5 conductivity of composites prepared by different coupling agent ratios

As can be seen from Figure 5, the best result was obtained when the ratio of 1:1 was used. The conductivity is $4.79 \times 10^{-4}$ S/m, an increase of 270% over the untreated. The electrical conductivity of the composites after surface treatment is higher than that of the untreated ones.

3.4.2 Effect of surface treatment of carbon fiber powder on impact strength of composites
The impact test results of the composite prepared by adding the treated carbon fiber powder are shown in Figure 6.
It is found from Figure 6 that the impact resistance of the material increases first and then decreases with the addition of coupling agent. The best condition is that the impact strength reaches 2.62kJ/m² when adding a part of treatment agent, which is 84.5% higher than that of untreated.

The results show that the surface properties of carbon fiber powder are improved, and the mechanical friction and interfacial adhesion of two phases (filler and resin) are enhanced, which has a good effect on the mechanical properties.

3.5 Testing and characterization

3.5.1 Infrared spectrum

Fourier transform infrared spectroscopy was used to analyze the carbon fiber powder before and after surface treatment. The test results are shown in Figure 7.

The carbon fiber powder used in the laboratory is micron scale, so it is not easy to combine with the resin, and it is easy to agglomerate. After the hydrolysis of KH-570, the groups formed dehydrate with O-H (contained on the surface of carbon fiber powder) to form covalent bond, which is more conducive to the interface bonding with the resin matrix.

3.5.2 Thermogravimetric analysis

There are four kinds of composite materials: blank epoxy resin, graphene/epoxy resin composite, carbon fiber powder/graphene/epoxy resin composite, and carbon fiber powder/graphene/epoxy resin composite with the best surface treatment. The results are shown in Figure 8.
Figure 8 thermogravimetric analysis

It can be seen from figure 8 that the composite material begins to decompose at 400℃. According to the data, the temperature of the composites increases when the weight loss is 10%, but there is no obvious change when the weight loss is 50%. At 800 ℃, the residual carbon content of the composites increases gradually, which is 18.1% higher than that of pure epoxy, 64.0% higher than that of pure epoxy, and 57.5% higher than that of pure epoxy. The results show that the addition of graphene and carbon fiber powder can effectively improve the thermal stability of the composites.

4. Conclusion

EP based conductive composites were prepared with carbon fiber powder and graphene as conductive fillers, which significantly improved the electrical conductivity and impact strength of EP. When the content of graphene is 10wt%, the conductivity of the composite increases from 0S/m to 1.396×10⁻⁵S/m, the impact strength is 23% higher than that of pure EP. When adding 10wt% graphene and 15wt% carbon fiber powder, the conductivity reaches 1.296×10⁻⁴S/m, which is 10 times higher than that of graphene (10wt%)/epoxy composite, and the impact strength is also increased by 7.6%. The electrical conductivity and impact strength of the composites prepared by silane coupling agent carbon fiber powder are 270% and 84.5% higher than those of the untreated ones, respectively.

Reference

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