

Preparation and characterization of graphite/thermosetting composites

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Abstract. Graphite/thermosetting composites were prepared via a melt blending and compression–curing process using epoxy and phenolic resins as polymer matrices. The flexural strength, flexural modulus and electrical conductivity of the composites were investigated. The composites containing 55 wt% graphite showed the maximum flexural strength and modulus. Scanning electron microscopy results showed that the thermosetting resins and graphite were uniformly dispersed on the fractured surface of the composites. The electrical conductivity of the composites increased with an increase in the graphite content. The flexural and electrical properties of the composites improved significantly by the addition of a carbon fibre cloth (CFC) or a CFC and carbon nanotubes.

Keywords. Graphite; carbon fibres; mechanical properties; electrical conductivity; carbon nanotubes.

1. Introduction

Grounding grids of power systems are used to eliminate the charge accumulated in electrical equipment. Such grids are buried directly into the ground and are usually built using metals. However, electrochemical corrosion can degrade grounding grids. The corrosion products formed on the surface of grounding grids affect their conductivity. The complete corrosion of grounding grids decreases their power supply capacity, thus endangering the safe operation of the equipment [1–4].

In China, flat steel is extensively used for grounding grids. Steel is cheaper and shows better thermal stability than copper. However, its corrosion resistance is poor. Galvanized steel has been used to improve the corrosion resistance of grounding grids. However, the galvanized layer becomes corroded in soil. Hence, the development of corrosion-resistant grounding grids is challenging [5–8].

Graphite has a layered structure with each carbon atom attached to three other carbon atoms by covalent bonds with the layers held together by van der Waals forces. Because each carbon atom in graphite emits a delocalized π electron which can move freely in the direction of the plane, graphite shows good electrical and thermal conductivities. Additionally, graphite shows excellent corrosion and thermal shock resistances. Therefore, graphite-based materials are widely used in conductive materials, microwave absorbing materials, electromagnetic shielding materials and antistatic materials [9,10].

Conventional sintering and compression sintering are the synthesis methods for graphite materials. Moulding involves the following steps: carbon powders or graphite powders and graphitized resin are mixed, moulded and graphitized at high temperature under a reducing atmosphere or vacuum. This method has some disadvantages such as high process temperature, energy consumption and cost [11].

Recently, *in situ* synthesis methods have been used for the preparation of graphite/thermosetting composites. In these methods, one or several reinforcements are formed in the matrix through *in situ* polymerization for strengthening the composites [12]. Among various thermosetting resins, epoxy and phenolic resins are widely used as polymer matrices for the preparation of conductive composites because of their excellent mechanical properties, thermal stability and solvent resistance. Epoxy and phenolic resins also have a high interface with graphite [13,14]. The resulting composites show corrosion resistance similar to that of graphite. Although the polymer resin content of these composites is low, they maintain the processability of polymers and can be moulded by typical polymer processing techniques such as extrusion, moulding and injection moulding. Therefore, graphite/thermosetting composites can be easily prepared on a large scale, and the production cost of the composites can be greatly reduced by one-time moulding [15].

In this study, graphite/thermosetting composites were prepared using epoxy and phenolic resins as polymer matrices via a melt blending and compression–curing process. The effect of the graphite content on the flexural strength,

flexural modulus and electrical properties of the composites was investigated. The effects of different fillers on the mechanical and electrical properties of the composites were also investigated.

2. Experimental

Diglycidylether of bisphenol A (E-51) (epoxide equivalent weight of 185–208 g per equivalent, Feicheng Deyuan Chemical Co. Ltd, China) was used as the epoxy resin in this study. The thermally latent initiator *N*-benzylpyrazinium hexafluoroantimonate (BPH) was synthesized using a method reported elsewhere [16]. Phenolic resin was prepared using a method reported previously. Graphite with a carbon content of 90% was supplied by Jilin Graphite Industry Co., China. The carbon fibre powders and carbon fibre cloth (CFC, K12, bidirectional fibres) were supplied by Jilin Jiyan High-Tech Fibers Co. Ltd. The diameter and length of the carbon fibre powders were 7 and 10–70 μm, respectively. Hydroxyl-functionalized multiwall carbon nanotubes (CNTs) with a diameter of 20–30 nm, length of 10–30 μm and OH content of 1.76 wt% (MH5 111216) were supplied by Chengdu Organic Chemicals Co. Ltd. of the Chinese Academy of Sciences.

Phenol (112.9 g, 1.2 mol), 37% formaldehyde (121.7 g, 1.5 mol) and ammonia (2.1 g, 0.06 mol) were added to a 500 ml four-necked round-bottomed flask equipped with a mechanical stirrer, thermometer sensor and reflux condenser. The mixture was heated slowly to 100°C and reacted for 2 h. The unreacted substance and water were removed by distillation at 80°C under vacuum. The phenolic resin was obtained as a viscous liquid (yield: 94%).

The DGEBA/phenolic resin ratio of the thermosetting resin was 50/50 wt%/wt%. The graphite content of the graphite/thermosetting composites was varied from 50 to 75 wt%. The graphite/thermosetting composites were prepared by a melt blending and compression-curing process. The desired amounts of DGEBA, phenolic resin, graphite and BPH were mixed in a mixer at 80°C for 1 h. The resulting mixture was injected into a preheated mould. The mould was compression-cured at temperatures ranging from 100 to 140°C and a pressure of 13.5 MPa for 3 h.

Graphite/thermosetting/CFC laminates were prepared as follows: the desired amounts of DGEBA, phenolic resin, graphite and BPH were mixed in a mixer at 80°C for 1 h. The mixture was injected into a preheated mould in which a specific amount of CFC was placed. The mould was compression-cured over the temperature range of 100–140°C at a pressure of 13.5 MPa for 3 h. The resulting laminates were sandwich-shaped and are shown in figure 1.

Flexural testing was performed using a three-point bending test according to the GB/T13465.2-92 standard. The specimen size was 10 × 10 × 64 mm³.

The resistivity of the composites was measured at room temperature using a DC resistance tester (AST10A) according

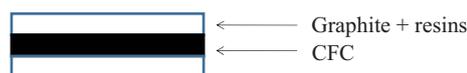


Figure 1. Structure of CFC/graphite/thermosetting laminates.

to the GB/T 24525-2009 standard. The sample size was 5 × 50 × 50 mm³.

The morphologies of the composites were investigated using field emission scanning electron microscopy (S-8000/Hitachi).

3. Results and discussion

The graphite/thermosetting composites were manufactured via a melt blending and compression-curing process. The mechanical properties of the composites were evaluated by determining their flexural strength and elastic modulus. The values of the flexural strength and elastic modulus were calculated using the following equations [17,18]:

$$F = \frac{3PL}{2bd^2}, \quad (1)$$

$$E_b = \frac{L^3}{4bd^3} \frac{\Delta p}{\Delta m}, \quad (2)$$

where F is the flexural strength, E_b the elastic modulus, P the applied load, L the span length, b the width of the specimen, d the thickness of the specimen, ΔP is the change in force in the linear portion of the load-deflection curve and Δm is the corresponding change in the deflection.

Figure 2 shows the flexural strength of the graphite/thermosetting composites as a function of the graphite content. The flexural strength of the neat resin system was 90.5 MPa,

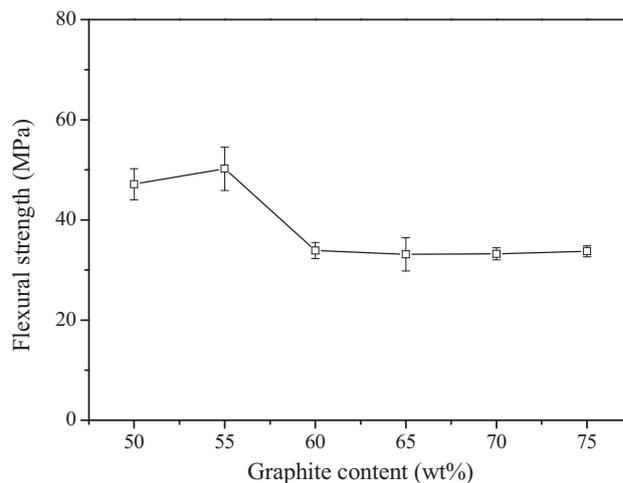


Figure 2. Flexural strength of graphite/thermosetting composites as a function of graphite content.

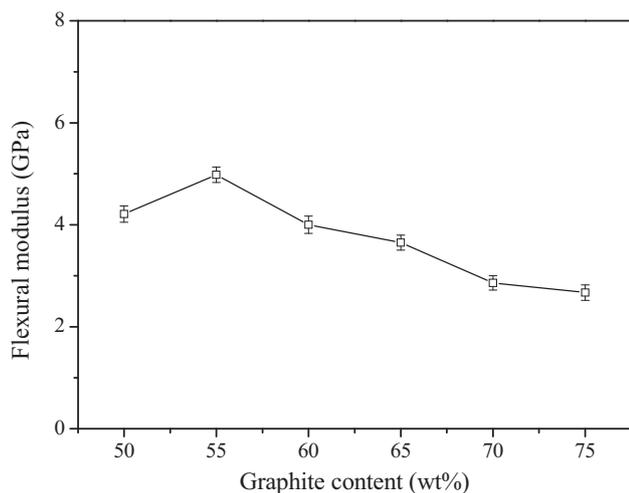


Figure 3. Flexural modulus of graphite/thermosetting composites as a function of graphite content.

whereas the flexural strength of the graphite/thermosetting composite containing 50 wt% graphite was 47.2 MPa, which was 48% lower than that of the neat resin system. The flexural strength of the composites increased as the graphite content was increased from 50 to 55 wt% and was the highest for the composite containing 55 wt% graphite. At the graphite content of 60–75 wt%, the flexural strength of the composites remained constant [19,20]. Thus, 75 wt% was chosen as the optimum graphite content for the preparation of the graphite/thermosetting composites using various fillers.

Figure 3 shows the relationship between the flexural modulus and graphite content of the graphite/thermosetting composites. The flexural modulus of the composites first increased with an increase in the graphite content (up to 55 wt%) and then decreased.

Following the flexural strength tests of the composites, their morphology was investigated using SEM. Figure 4 shows the SEM image of the composites after the flexural strength tests. The thermosetting resins (epoxy and phenolic resins) and graphite are uniformly dispersed on the fractured surface of the graphite/thermosetting composites [21,22].

The electrical properties of the graphite/thermosetting composites were evaluated by determining their electrical conductivity. The electrical conductivity was calculated as follows [23,24]:

$$\sigma = \frac{1}{\rho} = \frac{I \cdot L}{U \cdot S}, \quad (3)$$

where σ is the conductivity, ρ the resistivity, I the applied current, L the thickness of the sample, U the voltage through the sample and S the cross-sectional area of the sample.

Figure 5 shows the electrical properties of the graphite/thermosetting composites as a function of the graphite

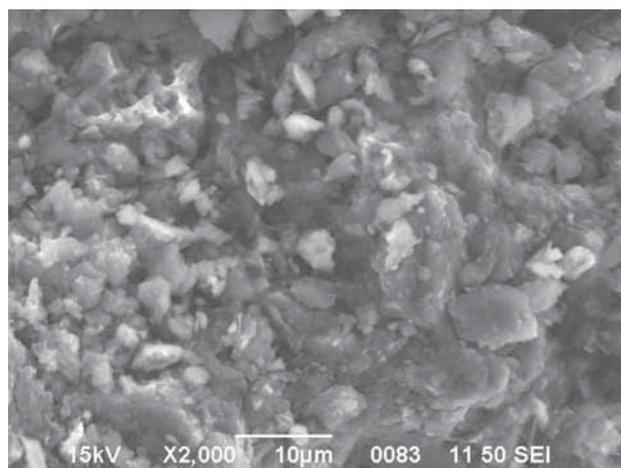


Figure 4. SEM image of graphite/thermosetting composites after flexural strength.

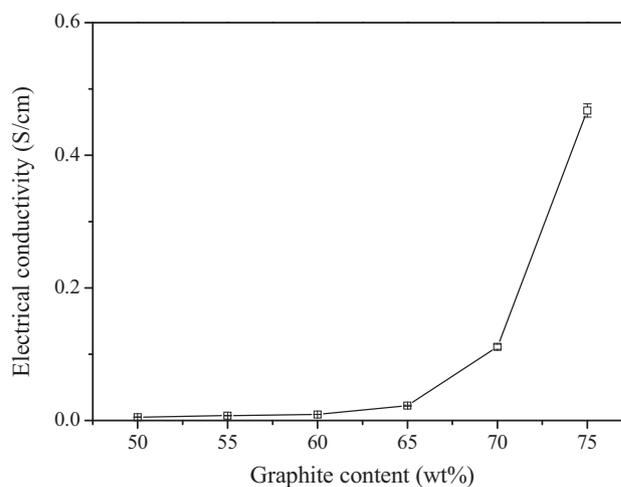


Figure 5. Electrical properties of graphite/thermosetting composites as a function of graphite content.

content. The electrical conductivity of the composites increased with an increase in the graphite content. At graphite contents higher than 65 wt%, the electrical conductivity of the composites increased significantly because of the good electrical conductivity of graphite [14,25].

Table 1 shows the flexural and electrical properties of the graphite/thermosetting composites prepared using various fillers. The flexural strength and flexural modulus of the composites increase slightly with the addition of the carbon fibre powders. The flexural strength and flexural modulus of the laminates improve by the addition of CFC or CFC and CNTs. These results can be attributed to the good interfacial adhesion between the CFC and the thermosetting resin formed by the reaction between the epoxy resin on the surface of the CFCs and the thermosetting matrix. This interfacial adhesion facilitated a load transfer

Table 1. Flexural and electrical properties of graphite/thermosetting composites with various fillers.

No.	Filler	Flexural strength (MPa)	Flexural modulus (GPa)	Electrical resistivity (S cm ⁻¹)
1	—	34.74 ± 1.12	2.67 ± 0.15	0.47 ± 0.01
2	1 wt% carbon fibre powders	35.38 ± 1.29	2.80 ± 0.04	25.97 ± 0.67
3	1 layer CFC	41.69 ± 1.32	3.00 ± 0.02	33.33 ± 0.89
4	1 layer CFC + 0.5 wt% CNTs	41.13 ± 1.42	3.19 ± 0.02	48.41 ± 1.43

Graphite content, 75 wt%; CFC, carbon fibre cloth; CNTs, carbon nanotubes.

from the matrix to the CFC, thus, efficiently absorbing the external energy to increase the flexural strength of the composites [26–29].

The electrical conductivity of the graphite/thermosetting composites increased significantly with the addition of the carbon fibre powders. A further increase was observed by the addition of CFC or CFC and CNTs. These results can be attributed to the high electrical conductivities of CFC (10³ S cm⁻¹) and CNTs (10⁴ S cm⁻¹) [30–33].

4. Conclusions

Graphite/thermosetting composites were prepared using a melt blending and compression–curing process and their mechanical and electrical properties were evaluated by measuring their flexural strength and flexural modulus and electrical conductivity, respectively. The composite with 55 wt% graphite showed the maximum flexural strength and flexural modulus of the composites. The flexural strength of the composites remained constant at the graphite contents of >60 wt%. The SEM result showed that the epoxy and phenolic resins and graphite were uniformly dispersed on the fractured surface of the graphite/thermosetting composites. The electrical conductivity of the composites increased with an increase in the graphite content from 50 to 65 wt%. The electrical conductivity of the composite increased significantly with an increase in the graphite content beyond 65 wt%. The flexural and electrical properties of the composites improved significantly by the addition of CFC or CFC and CNTs.

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