The effect of magnetic field intensity and treatment time on graphene / epoxy composites’ fracture toughness

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Abstract. The effect of the intensity of the magnetic field and the treatment time on the fracture toughness of graphene/epoxy composites is researched. Also, the mechanism of the effect of the magnetic field on the fracture toughness of graphene/epoxy composites and a method to improve the impact resistance is explored. Then, three-point bending tests are employed to characterize the fracture toughness of graphene/epoxy composite. The results show that the intervention of magnetic field could induce GNS to generated orientation arrangement, improving the fracture toughness of the graphene/epoxy composite. When the intensity of the magnetic field was increased, the growth rate of the fracture toughness slowed. However, when 2T magnetic was used to synthetically process the material, and when the processing time was less than 50 min, the fracture toughness of the composite material increased significantly.

1. Introduction

Epoxy resin is a high strength polymer material that is widely used in aerospace, machinery manufacturing, chemical building materials, shipping and other industries. Although epoxy resin has a high strength, it also has a lower impact resistance than other high-strength materials. In the epoxy resin toughening process, the preparation of composite materials, especially nano-composites, improve its impact resistance; this has become a hot topic for research. GNS sheets have thickness of a nanometer order. Also, the high modulus and high strength features of GNS make it an ideal component for the formation of nanocomposites [1]. Research related to GNS and epoxy composites has shown that that, in the case of direct recombination, the strength of the composite material is not increased significantly with an increase in the content of GNS [2,3]. In order to improve the strength of the composite material, the treatment of GNS must be modified. The intervention of a magnetic field can effectively change the orientation arrangement of GNS [4-15]. Based on the compound of the GNS and the epoxy resin, the toughening effect on the epoxy resin is studied. Toward this end, a constant magnetic field was used to intervene in the GNS and epoxy resin composite process. The orientation and arrangement of GNS in the epoxy matrix was changed by the magnetic field. This research can be used to deduce the effect of a magnetic field on composites.
2. Preparation and characterization method of the test material

2.1. Test material preparation
The GNS is dispersed in 100ml of acetone, at a frequency of 40KHz, for two hours in the power of 185W ultrasonic. After the GNS is uniformly dispersed, 6.6g epoxy is added, and the ultrasound is continued for two hours. The dispersed solution is stirred in a 75℃ environment by the use of a magnetic stirrer, then the solution is removed in acetone. While being stirred, the mixture is subjected to a degassing treatment for 30 min in order to remove any air bubbles. Finally, the degassed epoxy resin is poured into a homemade completed mold. After curing at 100℃ for twelve hours, the composite material is completely cured.

GNS / epoxy composites synthesis in a magnetic field is completed by using a constant magnetic field generated by an electromagnetic coil. Adjusting the magnetic field intensity to 0.5T, 1T, 1.5T, and 2T, the processing time is set at 10min, 30min, 50min, and 70min. After the magnetic treatment is completed, the mold is removed and placed in a curing oven, where it is cured for 12h at 100℃.

2.2. Test material characterization methods
The measure of fracture toughness is conducted with the ASTM D5045 for reference. The three-point bending method [16] is used. In order to reduce error, five splines from each sample are tested. Each sample is cut to length (S): Width (w): Thickness (B)=8:2:1. The prefabricated depth of sample surface is the 0.5W natural crack. Japan Shimadzu company’s AG-10TA electronic universal testing machine is adopted. The indenter radius is 2mm, the bearing fillet radius is 2mm, and the loading speed is 1mm/min. The Inspect. F scanning electron microscope of Dutch FEI company is adopted to observe the structure of the sample section.

3. Results and analysis of test

3.1. The effect of magnetic field intensity
As the composite materials were synthesized, magnetic fields of different intensities (0.5T, 1T, 1.5T and 2T) were exposed to the sample for 30 min. The composite sample which included GNS content of 1wt.% was acquired. The samples were then tested by three-point bending. The results of KIC and GIC are shown in figure 1.

![Figure 1. Relationship between $K_{IC}$, $G_{IC}$ and magnetic field intensity](image-url)
As can be seen from figure 1, as the strength of the magnetic field increased, KIC and GIC presented basically the same trend. When the magnetic field was 0.5T, KIC was 1.184MPam0.5. This is an 11.0% increase over the KIC without the presence of a magnetic field, 1.067MPam0.5. At the same time, the GIC value increased from 340.7J/m2 to 377.7J/m2, which is a rate of increase of about 10.9%. When these sets of data are contrasted, it can be clearly seen that the addition of a magnetic field significantly improved the fracture toughness of the composite. However, as the strength of the magnetic field continued to increase, the growth rate of KIC and GIC gradually slowed. When the magnetic field was enhanced 1T, 1.5T, or 2T, the growth of KIC and GIC was respectively 12.8%, 13.5%, 13.9% and12.3%, 13.5%, 13.8%. This implies that a lower magnetic field strength enables the GNS to achieve a better toughening effect. In contrast, the high intensity magnetic field only made a small increase of effect. The reason is that GNS has a good response for lower intensity magnetic fields, so that the GNS will have a certain orientation arrangement. The strength of the magnetic field can be enhanced to improve the degree of orientation of GNS; however, due to the restriction of the epoxy molecular chain, then its degree of orientation cannot raise too much, so the growth rate of the fracture toughness of the composite material gradually slows down.

3.2. The effect of treatment on time

In the process of composite material synthesis, the composite material was disposed of by using a 2T magnetic field for 10min, 30min, 50min, and 70min. The samples were tested to obtain the results shown in figure 2.

As can be seen from the figure, the fracture toughness values of KIC and GIC increased for the sample that was processed for 10min. Precisely, the increase was from 1.067MPam0 and 340.7J/m2 to 1.154MPam0.5 and 368.7J/m2. The growth rates are 7.5% and 8.2%. The sample that was processed for 30min also showed a similar growth trend. Although the slope efficiency of the curve indicates a slowing of the growth rate, the KIC and GIC values still appeared to increase significantly. However, when the time was extended to 50min and 70min, both values remained basically unchanged and the slope efficiency of the curve was approximately zero. This result could have been caused by two factors. First, the introduction of GNS could have increased the complexity and difficulty of the orientation. In a free state, due to the steric hindrance of epoxy molecular chains around GNS, the rotation of GNS becomes more difficult. As the degree of orientation with GNS improves, the epoxy molecular chain is closed to its entanglement, and GNS motion becomes more difficult. Therefore, even if the processing of the magnetic field is extended, GNS orientation movement does not proceed further. The second is to consider the epoxy curing time. The epoxy system solidified for 28min at 80℃ to fully achieve gelling. At this time, the viscosity of the system is very large, and the degree of crosslinking is already high. The system presents the basic curing characteristics. GNS is therefore fixed to the epoxy matrix, which can no longer produce orientation-induced motion.

![Figure 2. Relationship between K_{IC}, G_{IC} and magnetic field processing time](image-url)
3.3. Mechanism analysis of the impact of a magnetic field on the fracture toughness of graphene/epoxy composite material

The above test results indicate that treatment with a magnetic field can further improve the toughness of GNS/epoxy composites. This effect is due to GNS being formed into a magnetic field-induced orientation. The schematic principle can be described by figure 3.

![Figure 3. Schematic diagram of toughening mechanism in magnetic field](image)

When composites were processed in the presence of a magnetic field, because GNS itself is strongly magnetic, its orientation can be manipulated by a magnetic field. With the GNS arranged in the same direction, the nearby molecular chain of epoxy will create an orderly arrangement. A study of thermotropic liquid crystal toughened epoxy composites found that the orientation of liquid crystal molecules can be aligned in the direction of the external force when it is subjected to external force because of the mutual traction LCD and epoxy matrix, and the epoxy matrix is also present in the orientation of a certain trend. The regular arrangement of the epoxy molecules results in a greater amount of deformation in a parallel direction. When the composite material is subjected to external force, the epoxy molecular chain segments can deform to absorb the energy of the outside force, which reduces molecular chain fracture. At the same time, the high flexibility of GNS can also produce bending deformation and absorb some of the energy. This creates a synergistic effect of GNS and the epoxy molecular chain, so that material damage caused by an external force decreases; this achieves the goal of improving toughness. According to the theory of polymer mechanics, the increase of fracture toughness must be accompanied by changes of breakage elongation. From the tensile property tests for the above two samples, the changes of breakage elongation are shown in figure 4. As can be seen from the figure, the breakage elongation with the trend of the magnetic field strength and magnetic treatment time was consistent with the KIC and GIC. This confirms that the orientation of GNS in composites by use of a magnetic field produces a toughening effect.

![Figure 4. Relationship between magnetic field and elongation](image)
3.4. Electron microscopy analysis of graphene/epoxy composites with magnetic field

The most direct and effective method of characterizing samples is to use an electronic microscope to directly observe them. Figure 5 shows images of two samples, one treated with a magnetic field and one not.

![Figure 5. SEM images of different samples (a)untreated (b)1T](image)

The SEM image shows that the sheet of sample (a) without the treatment of magnetic field is arranged in a disorderly fashion, and the layered stacked state appears irregular. However, the treatment sample of 1T magnetic field induced a clear orientation structure, and the sheet was orderly arranged in the same directions. This indicates that the orientation of GNS was induced by the magnetic field. By observation of the fracture surface, it becomes clear that the orientation of GNS caused the ordered ethylene molecule segments. In contrast with figure 4, stress concentration points in figure 5(a) become obvious from observing both 5(a) and 5(b), which may be caused by maldistribution of GNS in the matrix. However, in figure 5(b) stress concentration points are rare. This indicates that the magnetic field promotes proper distribution of GNS in the matrix. It was also shown that the magnetic field is the reason behind the improvement of characteristics of the composites.

4. Conclusion

In the synthesis process of the GSN/epoxy composite, the magnetic field was shown to induce directional arrangement of GSN. Within a certain range, the degree of orientation will increase as the strength and length of the magnetic field also increase.

Based on the above testing results, the magnetic field is an effective method to improve the performance of GSN/epoxy composites.

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