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Microwave curing of carbon nanotube/epoxy adhesives

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ABSTRACT

In this paper we have shown that using microwave to cure epoxy adhesive containing multi-wall carbon nanotubes (MWNTs) not only leads to large time and energy savings, but also results in much higher strength. The well-dispersed MWNTs in the epoxy resin absorb microwave energy, acting as heat sources to cure the epoxy resin uniformly and effectively. Microwave curing can diminish the deterioration effect of lowering cross-link reaction by MWNTs as observed in conventional heating. Thus, as much as three times addition of MWNTs in the epoxy is allowed for optimum strength. As a result, 56% increase of the shear strength was observed when the epoxy adhesive doped with 3 wt% MWNTs was cured by microwave heating.

1. Introduction

In joining or repairing engineering structures, polymer adhesives are commonly employed. Among them, high strength epoxy adhesives that need to be cured at high temperature are widely used because they offer a number of advantages. However, there is continuous demand for shortening the time-consuming curing processes and increasing the strength of adhesives to improve their overall bonding performance.

Conventionally, the heat is applied to cure the epoxy adhesives by using furnaces, heating blankets or lamps. The conventional heating means are inefficient since a large part of the heat is lost to the surroundings. In contrast, microwave radiation provides an effective, rapid and localized heating. Hence microwave heating offers significantly greater energy efficiency and time saving [1]. However, the heating mechanisms associated with microwave are based on the dielectric properties of the materials to be heated [2]. Since neat epoxy is not efficient in absorbing microwave energy, to facilitate heating of the epoxy adhesive through microwave radiation, microwave energy absorbing agents such as magnetic particles, carbon black, metal particles, or metal fibers, were added to the adhesives [3,4]. Recent study has shown that in addition to strengthening composite materials, CNTs are also very good microwave energy absorbing agents [5–7].

The properties of CNT contained epoxy composites as adhesives have been widely studied. The CNTs added in polymer can bridge cracks and increase the elastic modulus, tensile strengths, hardness and tensile impact strength of the material [8–11]. The tensile modulus was observed to increase from 3.1 GPa to 3.71 GPa with 5 wt% CNTs added in MWNTs–epoxy composites [12]. Fracture toughness of the adhesives can be improved with appropriate content of CNTs added in the adhesives for use in joining steel, aluminum and composite materials [13,14]. With high content of CNT added in epoxy, both tensile strength and damping properties are improved [15].

Imholt et al. showed that CNTs have strong microwave absorbing properties [5]. Wang et al used microwave heating to transplant MWNTs onto different polymer substrates to fabricate flexible field emitter [6]. They demonstrated that by using microwave heating, polymer substrates can be joined with MWNTs in seconds [7].

Fotiou et al. [16] demonstrated that when epoxy mixed with CNTs was cured by microwave heating, at least 40% energy spent on curing could be saved. However, they noticed that CNT would agglomerate even with only 0.5 and 1 wt% CNTs addition in the epoxy prepared in their investigation. They commented that due to the existence of agglomerations, thermal spots has been created through microwave heating that result in local degradation of resin and so the stress to failure was eventually lowered with the addition of CNTs. Therefore, more studies are needed to apply microwave heating for producing advanced composite structures.

The present study reports the optimum processing schedules of the carbon nanotube contained epoxy composite (MWNTs/epoxy) adhesives that can be fast and uniformly cured by microwave radiation and provide large improvements in their mechanical properties.

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2. Material and methods

The carbon nanotubes used in this research were multi-walled carbon nanotubes with an average length of 5.3 μm. The epoxy adhesive contains DGEBA type epoxy resin, curing agent, and catalyst. The curing schedule suggested by the epoxy supplier is 150 °C for 30 min. It is found that the mixture of the epoxy adhesive with MWNTs can be stored for over two months at room temperature without any noticeable reaction.

The adhesive mixtures containing 0.5 wt%, 1 wt%, 2 wt%, and 3 wt% MWNTs were shear mixed by going through a three-roll mill five times to ensure that the MWNTs are well dispersed in the epoxy resin. The collected MWNTs/epoxy adhesives were degassed in a vacuum chamber before being used for joining application. A differential scanning calorimetry (DSC) was used to study the curing process of the adhesives. The materials to be joined by various kinds of adhesive in the research are cross plied laminates glass fiber reinforced polymer (GFRP) plate with a tensile strength of 375 MPa, which are commercially available. Before bonding, the surfaces of the GFRP laminates were roughened by P240 grit size emery paper and wiped by acetone soaked napkin.

A single mode microwave heating system [17] which has a fixed uniform heating region was employed in the study. By adjusting the microwave power, the temperature of the sample placed in the uniform heating region can be controlled. In microwave heating experiment, the temperature was measured by an IR pyrometer. Fig. 1(a) is a schema of the specimen assembly for microwave heating. A black GFRP bar was fixed over the samples to minimize the effect of specimen color on the accuracy of the IR measurements. The IR pyrometer measured the temperature of the black GFRP surface, and the corresponding temperature of the adhesive joints was calibrated in a preliminary study with a thermal couple attached to the adhesive, as illustrated in Fig. 1(b).

3. Results and discussion

3.1. Curing of the adhesives by conventional and microwave heating

The onset of the epoxy adhesive's curing reaction happened at 134 °C as measured by DSC analysis. To more precisely determine the curing time of the adhesives, adhesives with a thickness of 110 μm were coated on GFRP substrates and subsequently cured by either conventional and microwave heating at 150 °C or 170 °C for different times up to 60 min, long enough to assure the complete curing of the adhesive specimens. We found that the completely cured specimens have a stable hardness of HV 20.3. The time needed for complete curing was then determined when the Vicker's hardness of the specimens reached the stable value HV 20.3, indicating that the specimen was definitely complete cured.

The present study showed that curing of adhesive through conventional heating was completed in 25 and 20 min at 150 °C and 170 °C respectively. In contrast, only 8 min and 5 min were needed by microwave heating at 150 °C and 170 °C, respectively. The curing time by microwave heating was shortened to just 1/3 – 1/4 of that required by conventional heating means. The saving of curing time is attributed to that the well dispersed MWNTs could absorb microwave energy to act as the heating sources in the adhesive, leading to a more uniform volumetric heating.

Curing at a higher temperature can further shorten the curing time. However, this would be limited by the decompose temperature of the polymer. Therefore, it is impractical to shorten the curing time through increase of temperature beyond 170 °C. In the following work, curing of the adhesives was all performed at 170 °C either by conventional heating for 25 min or by microwave heating for 8 min.

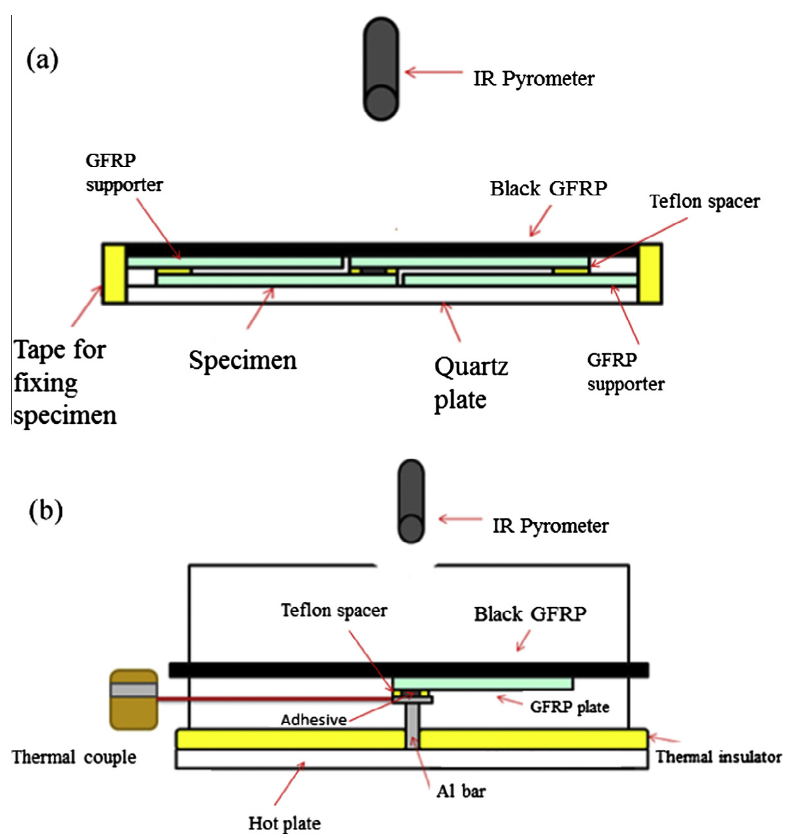

Fig. 1. (a) The specimen assembly for microwave heating and (b) experiment setup for calibrating the temperature measuring of IR pyrometer.