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Environmental durability of glass fiber epoxy composites filled with core–shell polymer particles

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ABSTRACT

A manufacturing technique was successfully adopted for incorporating core–shell polymer (CSP) particles into glass fiber reinforced polymer (GFRP) composites. CSP particles were firstly added into inter-ply interfaces during the lamination process. Conventional as well as CSP particle filled GFRP laminates were then cured at 122 °C under high pressure while in a vacuum environment. Good adhesion between the particles and the epoxy matrix was achieved during the curing. GFRP laminates with or without particles were immersed in water at 80 °C for different durations. These conditioned laminates were later subjected to low-velocity impact tests. It was found that the GFRP laminates with CSP particles absorbed more moisture while had less structural defects formed during the conditioning. The CSP particles significantly improved the impact properties of the GFRP laminates. After hygrothermal aging, the beneficial effects brought about by these particles for effectively resisting impact loads diminished. The underlying mechanisms were identified, studied and explained.

1. Introduction

Glass fiber reinforced polymer composites are among the most widely used composite materials. These composites are prepared by incorporating high-strength fiberglass into a polymeric matrix such as an epoxy resin. The advantages of GFRP composites include high strength-to-weight ratios [1–2], lower costs as compared with carbon fiber reinforced composites [3], and good chemical resistance [4]. Nowadays, GFRP composites are widely used as structural materials on aircrafts [5], automobiles [6], and ships [7], as well as in other industrial segments such as wind energy industry [8] and construction industry [9].

Fiber reinforced polymer (FRP) composites are usually susceptible to impact damages which in many cases are externally invisible while harmful to the residual strength of these materials [10–12]. Many techniques have been attempted by researchers to improve the impact performance of composite materials. These techniques can generally be divided into two categories, namely, fiber related and matrix related. Hybrid composites which have two or more types of fibers embedding in single matrix materials [13], translaminar reinforcements such as stitching [14] and surface modification of glass fibers such as treatment with silane coupling agents [15] are fiber related solutions aiming at improving the impact properties of composites. Matrix related techniques available in the literature include but are not limited to application of tough thermoplastic resins [16], addition of fillers into matrix resin such as nanoclay [17] and carbon nanotubes [18].

There are, however, limitations associated with each of the techniques listed above, such as complexities in design and manufacturing, increased tooling costs, deterioration in other mechanical properties of the composites. One promising solution for the toughening of composite materials is a particle-based inter-ply technique [19]. Ali and Joshi [20] applied core–shell polymer particles onto the inter-ply interfaces of GFRP composites. Due to the incorporation of CSP particles, the peak contact force under low-velocity impact increased while the extent of impact damage decreased dramatically. However, in their investigation the laminates were cured in a convention oven rather than in an autoclave. Autoclave curing is widely accepted in the industry as it provides good control of temperature, pressure and vacuum. Further investigation on autoclave cured laminates is necessary to explore the impact performance of laminates filled with CSP particles.

Another disadvantage of polymer matrix composites (PMCs) is their susceptibility to the influences of hygrothermal environments. Owing to the heterogeneity of PMCs microstructures, the polymer matrix usually absorbs moisture under humid conditions, and the coatings or treatments on the fibers are possible factors that make PMCs vulnerable to hygrothermal environments. The environmental durability of PMCs has attracted attention from both the academia and the industry for several decades. In most cases, the various properties of PMCs were negatively affected after exposure to hygrothermal environments [21–22].

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These include the glass transition temperature of the matrix resin [23], compressive strength [24], flexural properties [25], and so on [26]. Positive effects of moisture and hygrothermal aging were also reported. Zhong and Joshi [27] carried out water immersion aging on carbon fiber reinforced composites. It was found that the peak contact force under low-velocity impact increased by 16.45% after 48 days of aging (at 80 °C). The interaction between composites and hygrothermal conditions is a rather complex phenomenon. It depends on the particular combination of matrix and reinforcement, the service environments and the duration of exposure. More research efforts are still needed in this area.

Hygrothermal environments are, in many cases, inevitable for composite structures in service. Thorough understanding of the various aspects of composites filled with CSP particles is necessary before using these particles on real composite structures. To the best of the authors' knowledge, no research work concerning the effect of moisture on CSP particles is currently available in the literature. In this investigation, the emphasis was placed on the environmental durability of CSP particles and GFRP composites filled with these particles. The first step was to incorporate CSP micro particles into GFRP laminates through autoclave curing. GFRP laminates thus prepared were immersed in water at 80 °C for hygrothermal aging. Water immersion aging provided hygrothermal environment while accelerated the moisture ingress into the composite materials. The temperature of 80 °C is close to the possible highest temperature of a composite structure or a hot component on an aircraft or an automobile, especially under direct sun light. Low-velocity impact test was carried out on these hygrothermally aged laminates. A detailed experimental plan was designed and carried out to explore the environmental durability of CSP particles and their influences on the impact performance of GFRP composites. Tensile test was conducted to check the effects of hygrothermal exposure on the strength of GFRP composites.

2. Experimental details

2.1. Sample preparation

Both samples with and without CSP particles were fabricated using L-530 (7781) prepreg materials, which were manufactured by J. D. Lincoln Inc. The recommended curing temperature for this prepreg material is between 121–135 °C. The epoxy resin (L-530), which constituted 38 wt.% of this prepreg material, was diglycidyl ether of bisphenol A (DGEBA). The reinforcement used in this prepreg material is 7781 fiberglass fabric. Pregregs of L-530 (7781), which were previously stored in a freezer at −30 °C, were cut into laminae of 100 mm × 100 mm after being defrosted in lab condition for 4–5 h. Later, laminae were stacked into laminates of 8 plies for curing.

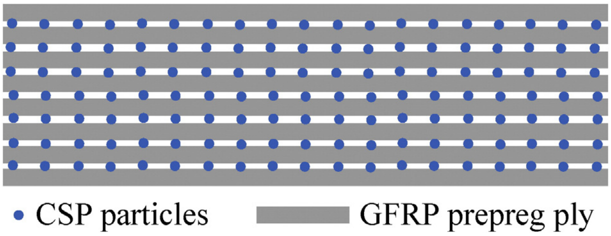


Fig. 2. Schematic representation of a GFRP laminate (before curing) with CSP particles dispersed at all the 7 interfaces.

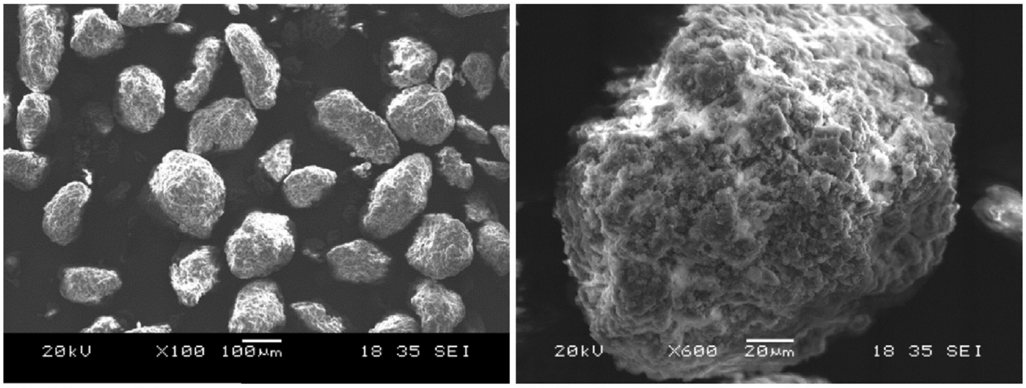
The CSP particles (PARALOID EXL-2314) used as impact modifiers for GFRP laminates were supplied by the Dow Chemical Company, Singapore. These particles were in the form of a fine white powder and appeared as discrete particles without any agglomeration. They have a soft rubber (PBA) core and a transparent shell of poly (methyl methacrylate) (PMMA) with an epoxy functional group grafted to the shell to improve the interfacial bond of the particles with epoxy resin. The morphology of as received CSP particles is shown in Fig. 1.

During the fabrication of the GFRP laminates with embedded CSP particles, CSP particles were dispersed manually at all the 7 inter-ply interfaces as shown schematically in Fig. 2. The lamination process for the GFRP laminates with the CSP particles is shown in Fig. 3. The CSP particles were weighted using an analytical balance. In order to achieve a more uniform distribution of CSP particles at the interfaces of the GFRP laminates, the CSP particles were firstly dispersed onto a peel ply (Teflon release film) at an areal density of 30 g/m^2 (Fig. 3). Later, the CSP particles on the peel ply were successfully transferred to the GFRP prepreg ply by placing the GFRP prepreg ply on the top of the peel ply. Due to the sticky nature of the GFRP prepreg, these particles easily stuck to it.

Both types of GFRP laminates — with and without CSP particles, were cured in the autoclave using the same curing program. The temperature and pressure applied during curing was 122 °C and 606.7 kPa, respectively. The final dwelling time (curing time) was 2 h. Detailed information about the cured GFRP laminates is summarized in Table 1.

2.2. Hygrothermal aging

In this investigation, during the hygrothermal aging samples were immersed in tap water at 80 °C in a general water bath. Therefore, the GFRP composites were exposed to the combined attack of elevated temperature and moisture. The temperature stability in the water bath is ±0.3 °C. The weight of the samples was measured using an analytical



(a) CSP particles (b) Surface of a CSP particle

Fig. 1. Morphology of as received CSP particles observed using SEM.