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Hybrid enhancements by polydopamine and nanosilica on carbon fibre reinforced polymer laminates under marine environment

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ABSTRACT

In this study, two enhancement methods, i.e., toughen the epoxy matrix by commercially available nanosilica and enhance the interfaces of fibres and matrix by autoxidation of dopamine were applied together in carbon fibre reinforced polymer laminates with potential large-scale applicability. Significant enhancements were found for Mode I interlaminar fracture toughness and interlaminar shear strength with the combined addition of nanosilica and polydopamine in the laminates. The enhancement mechanism is proposed as well. Salt spray tests were applied in this study to simulate a marine environment for the laminates. Model I interlaminar fracture toughness and interlaminar shear strength both decreased under the simulated marine environment with an increase in immersion time, but the deterioration was significantly mitigated when nanosilica and polydopamine were added together with still much higher mechanical properties measured after 3 weeks of salt spray immersion than in neat laminate without salt spray immersion, providing promising evidence for maritime engineering applications of such laminates.

1. Introduction

Growing demand exists for carbon fibre reinforced polymer (CFRP) composite materials with enhanced properties, which are essential for applications in engineering fields, especially in harsh environments. As CFRP is a combination of fibre and matrix, its properties are dominated by high strength and stiffness in fibres as well as low strength in the ductile polymer matrix and the interfaces between fibres and matrix. These poor polymer properties and weak interfaces between fibres and polymer significantly limit the applications of CFRP. Consequently, it is of interest to enhance the polymer resin as well as the interfaces between the resin and fibre of the CFRP composites to extend laminate applications in various fields.

Significant work has been done to enhance polymer matrix as well as interfaces in recently years, with some studies focusing on large-scale production and commercialization. The use of nanoparticles to toughen polymer matrix is one promising method, employing such particles as nanosilica, halloysite nanotubes, and carbonaceous nanoparticles such as graphene nanoplatelets and graphene oxide. Improvements in Young's modulus, fracture toughness and tensile strength have been reported [1–6]. However, one of the challenges in using nanoparticles is the achievement of homogeneous dispersion of high weight/volume ratio nanoparticles in matrix while maintaining comparatively low viscosity. The difficulty is that viscous resin systems cannot easily impregnate continuous fibres or fibre fabric during CFRP production. Meanwhile, based on a resin infusion process, the filtering of dense fibre bundles against agglomerated nanofillers can lead to severe segregation and depletion of nanofillers in matrices [7,8], also significantly offsetting the enhancement effects of nanoparticles in laminates. One of the solutions is the use of *in situ* synthesized methods such as the sol-gel manufacturing process, whereby particle size and excellent distribution are unaffected during any further processes. Several commercially produced nanosilica-modified epoxies are available, such as Nanopox F400, a concentration of 40 wt% nanosilica in diglycidyl ether of bisphenol A (DGEBA) epoxy resin [9], with an average particle size of 20 nm and a narrow range of particle size distribution. In our previous research, only 2 wt% nanosilica added in the epoxy improved the fracture toughness and corrosion rate under a marine environment [9]. Meanwhile Sprenger reported using nanoparticles no longer further improvement the delamination fracture value when the value reached to 500 J/m^2. Above that value, the dominated delamination fracture of laminate often changes from matrix failure to interfacial failure [10]. Therefore, the addition of a single type of nanoparticle did not further mitigate the occurrence of delamination fracture.

Sizing is a method of wetting out fibre surfaces to improve the poor interfacial adhesion of CFRP. Polydopamine (PDA) is a bionic material

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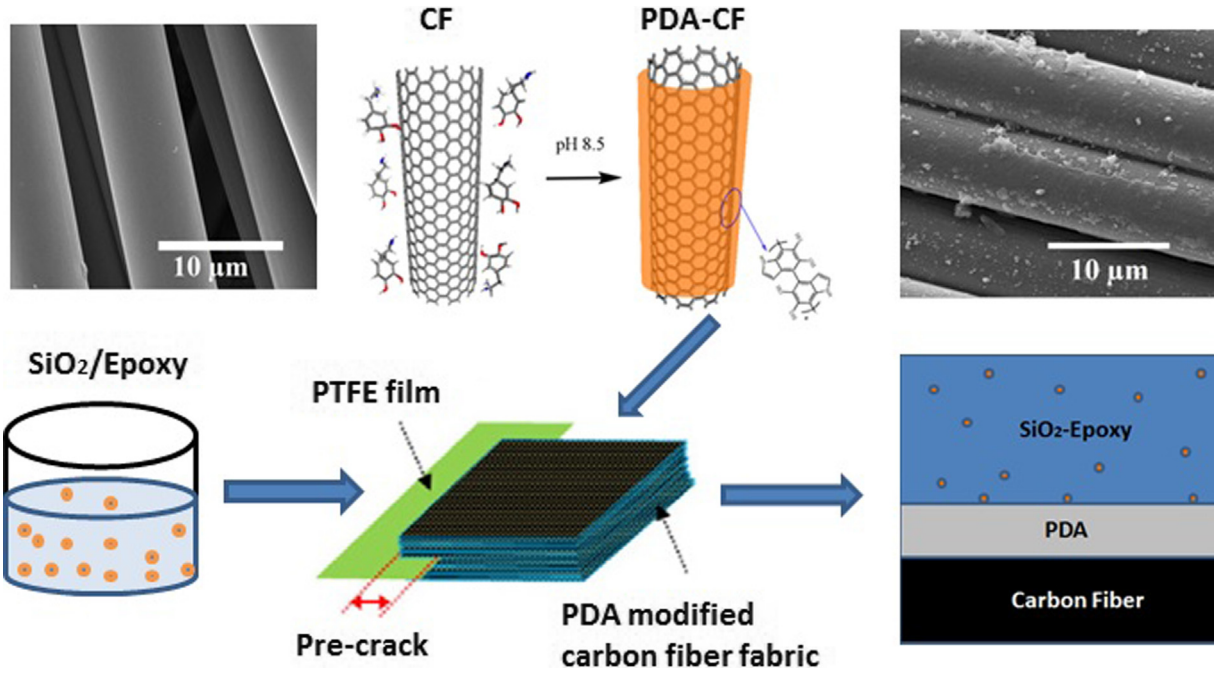

Fig. 1. Schematic drawing demonstrating the hybrid enhancement strategy by toughening epoxy matrix with nanosilica and enhancing the interface by applying polydopamine on carbon fibre surfaces. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

which has excellent adhesion to a range of solid surfaces, such as metals, oxides, polymers and ceramics by autoxidation of dopamine in basic aqueous solutions [11]. Recently, it has also been applied to modify nanoparticles such as carbon nanotubes [12], graphene [13] and clay [14], revealing excellent ability to improve the mechanical, thermal and electromagnetic interference shielding performance of polymer matrices. In addition, PDA has been used to modify short carbon fibres, revealing significant improvement in tensile strength and Young's modulus [12]. Furthermore, Yang et al. reported that the catechol groups in PDA were forming hydrogen bonds with polar groups in epoxy [15]. In addition, the interfacial covalent bonding was forming between PDA and epoxy because of the primary and secondary amine groups in PDA may react with epoxy groups and the amine hardener may react with PDA [16,17]. We reported using a simple method for surface modification to improve the load transfer between carbon fibre and epoxy matrix, increase the fractured interface friction and reduce unstable crack growth in CFRP composites [18].

The application of CFRP composites in maritime engineering was initially the demand of building lightweight, strong, corrosion-resistant durable naval vessels. CFRP can overcome corrosion problems experienced with steel or aluminium alloys and environmental degradation suffered by wood. Additionally, CFRP can significantly reduce the weight of a structure but still maintain the desired performance and structural integrity. However, under the marine environment, the mechanical properties of laminates degrade due to UV, moisture, temperature and ageing – creating the potential of accidentally creating fracture. Bastioli et al. reported water aging may strongly affect the matrix behaviour, by producing changes in its chemical and physical nature by itself or in conjunction with other chemical or physical agents such as heat and ultraviolet light [19,20]. Moreover, the fibre/matrix interface can be degraded by a hydrolysis reaction of unsaturated groups within the resin under marine conditions [21–23]. A concern is the incomplete understanding and shortage database of using fibre reinforced composites as marine structures with long-term durability. Consequently, to efficiently enhance the properties of CFRP and extend its applications in the harsh marine environment, there is interest in improving the mechanical performance at least to offset the deterioration generated by that environment.

Current enhancements to interfacial adhesion rely on sizing wet-out fibre surfaces. However, the inherent poor properties of polymer matrix render these solutions inefficient. Furthermore, polymer matrix enhancement can only toughen the matrix itself, with less enhancement of the interfaces. In this work, we demonstrate a feasible hybrid method to enhance the laminate, with potential large-scale application under marine environments. Commercially available nanosilica can provide the necessary polymer matrix toughness, while polydopamine on carbon fibre surfaces can provide significant interfacial adhesion among fibres and matrix. With those hybrid enhancements, laminates can offset deterioration under a simulated marine environment and still achieve superior mechanical performance to that displayed by neat laminate without salt water immersion.

2. Experiments

2.1. Polydopamine for CFRP interfacial enhancement

The as-received carbon fabric was submerged in acetone for 48 h to wash off the commercial sizing and impurity. For fabrication of the PDA sizing fibres, 4 g dopamine hydrochloride (Sigma, Australia) was dissolved in a mixed solution of deionized water (4000 mL) and aqueous solution of TRIS (3.6 g tris(hydroxymethyl)aminomethane, 1000 mL deionized water), with magnetic stirring for 30 min. 200 gm unidirectional carbon fibre fabrics (Hexcel, USA) were prepared into 8 layers with the size of 30 cm * 30 cm then placed in a container and the mixed solution was transferred to the container. The container was shaken by a benchtop orbital shaker (Labec, Australia) at 100 rpm for 24 h at ambient temperature. Then, the modified carbon fibre fabrics were collected, washed with deionized water several times to remove the residual dopamine, and dried in a vacuum oven at 40 °C for 24 h. Finally, the thickness of PDA layer on the carbon fabric was 50–100 nm. From our previous report, approximately 3.2 wt% of polydopamine was coated on the carbon fabric [20].

2.2. PDA-SiO_2-CFRP composites preparation

A vacuum-assisted resin transfer moulding (VARTM) process was