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Hydrolytic stability of PC/GF composites with engineered interphase of varying elastic modulus

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Abstract

Effects of elastic moduli of the interphase on the hydrolytic stability and mechanical properties of polycarbonate (PC) reinforced with unidirectional E-glass and S2-glass fibers was investigated. Interphase, approximately 100 nm thick, were deposited onto E- and S2-glass fibers using various deposition techniques. Elastic modulus, E_i, of the interphase was determined using two experimental techniques, i.e., measurement of Rayleigh wave speed and vibrating piezoelectric crystal. In this work, E_i varied from 0.1 to 6 GPa. Single-embedded fiber fragmentation test was utilized to determine the average shear strength of the interphase, τ_a. Increasing E_i resulted in an increase of τ_a from 19 MPa for the soft interphase to 42 MPa for the rigid interphase, respectively. It was also found that the usual silane sizing agents used commercially to promote matrix–fiber bonding led to a reduction of hydrolytic stability of the interphace under extreme conditions of stress and moisture. Elastic modulus and strength of the laminate were measured in the direction parallel and perpendicular to the direction of fibers. Properties of both model single-fiber composites and 8-ply UD laminates were measured in the dry state and after exposure to 85 °C water for 100 h followed by 100 h drying at 100 °C. No significant reduction in longitudinal properties of the laminates resulted from the exposure to water. In some cases, significant reduction in transverse properties, controlled by the matrix and the interphase behavior, was observed. PC/bare fiber composite annealed at 275 °C for 10 min prior the immersion in water exhibited excellent retention of properties after long-term immersion in hot water. Similar results were found for composites with fibers treated with PC oligomer and plasma deposited MPTCS. This finding was attributed to the formation of a dense, high modulus interphase consisting of the deposited layers and immobilized PC chains near the fiber surface. In addition, improved wetting of these fibers by the PC contributed to less defective and, hence, more stable interphase.

Keywords: Composites; Mechanical properties; Hydrolytic stability; Polycarbonate; Interphase

1. Introduction

Over the last 40 years there has been a substantial amount of work published on the use of fiber reinforced composites (FRCs) in biomedical applications [1–4]. There has also been great interest to use FRC components in automotive industry [5]. Most of the FRCs investigated were based on thermosetting resin matrices. Short shelf life and the expensive production technology, however, limit the applicability of termoset-based composites for repair or replacement of corroded body parts in larger vehicles such as trucks and military transport vehicles, especially, since low volume of these parts is usually desired.

Among the thermoplastic FRCs used in automotive industry, glass mat thermoplastics (GMTs) gained the most commercial success. Excellent corrosion resistance and sufficient toughness led to extensive utilization of GMT materials. Some passenger car makers use glass mat reinforced thermoplastics (GMT) for bumper components, gear and clutch boxes, battery holders and other under the hood parts as well as in lower body parts. In addition, there is an increasing market for use of GMT in small trucks in applications such as car-

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go boxes, cargo beds, etc. The GMT still requires to employ expensive tooling and equipment. Use of thermoplastic matrices in long-fiber reinforced composites offers, in addition to low weight and durability, indefinite shelf life of semifinished parts, potential for relatively inexpensive tooling for low volume parts, possibility for heat forming of semi-finished parts and enhanced toughness compared to the thermoset-based FRCs. Easy recycling is another advantage of these materials. Relatively poor mechanical properties of the semi-finished GMT sheets, relatively large variations in mechanical properties given by the nature of the nonwoven reinforcement and inability to achieve the required automotive finish were the major shortcomings of commercially available GMTs [5].

While the only concern about the time dependence of mechanical properties of metals in the moist environment is their corrosion resistance, both time and moist environment can affect significantly the performance of FRC components, especially in load bearing applications. These effects in long-fiber thermoplastic composites are assumed to be mostly due to the hydrolytic deterioration of the fibers, matrix and interface [6] as well as the viscoelastic character of the matrix [7]. In the case of metal parts, there is little concern about the limits of applicable loads, since the material properties greatly exceed the actual requirements. The attainable mechanical properties of FRC parts, however, are much closer to the actual needs. The anisotropic character of FRCs is another factor which has to be taken into consideration. Since the transverse properties of unidirectional composites are generally poor, the triaxial stresses, inevitably created near defects such as fiber breaks could result in damage accumulation and, eventually, lead to premature failure of the part. Since transverse properties are matrix controlled, the susceptibility of matrix and matrix/fiber interface to the hydrolytic deterioration will be of crucial importance for the stability of the FRC parts in wet and aggressive environments. Since fiber reinforced composites can fail in several different modes [8], the identification of the possible failure mechanisms and the effect of moisture on these properties are of critical importance [9]. While the properties of FRC often recover from the effect of water upon drying, chemical changes in the matrix or interface and/or degradation of the fibers under stress can lead to a non-recoverable deterioration of the material in adverse environments [10,11].

Penetration of water into a composite can occur by both diffusion through the matrix bulk and capillary flow along the weak fiber–matrix interface. The permeation rate due to activated diffusion increases exponentially with temperature and is pressure independent. Capillary flow through defects (voids, crazes, etc.) and along non-bonded interfaces is pressure dependent [11]. The rate of water penetration along the interface is about 450 times more rapid than through the resin bulk.

Polymers containing ester groups (PC, PETG) have the propensity for undergoing chain scission by hydrolysis resulting in degradation of resin properties [7]. Hydrolysis of these polymers is accompanied by a reduction of molecular weight and viscosity and an increase in concentration of –COOH groups (PETG). The presence of glass fibers results in a leaching out of alkaline species during the water immersion which may catalyze the hydrolysis. Glass fibers may also degrade during water immersion, particularly if exposed while under stress [12–14]. In addition, most of the engineering polyesters exist as amorphous solids and the diffusion through amorphous phase is substantially faster than through a crystalline phase, under the same external conditions.

The pivotal effect of the interphase region on the performance of long-fiber thermoplastic composites is generally accepted. With the demand for new demanding applications of FRCs in various industries, the simple silane surface treatment of fibers becomes unsatisfactory and often limits the use of FRCs, especially, in moist environments. Various agents as well as deposition techniques have been used to alter properties of the interphase region in a controlled manner. It has been found that varying the deposition technique could result in interphase layers with varying elastic modulus which was shown to control the stress transferability to a great extent.

Since the E-glass fibers are the most widely utilized reinforcements for GMTs, these fibers were chosen as a reference reinforcement in this investigation. In addition, S2-glass fibers with enhanced elastic modulus, strength and thermal resistance were selected as alternative reinforcements useful for applications with higher added value. In this study, the effect of moisture on the flexural properties of polycarbonate reinforced with unidirectionally aligned E-glass and S2-glass fibers for car and truct body part applications was investigated. The principal goal of this study was to investigate the resistance to hydrolysis of the interface in unidirectionally oriented glass fiber reinforced model thermoplastic composites. In addition, investigation was performed to elucidate the effect of moisture on the flexural modulus, strength and ultimate strain of thermoplastic composites reinforced with unidirectionally aligned fibers, in the directions parallel and perpendicular to the fiber orientation.

2. Experimental

2.1. Sample preparation

Unidirectional E-glass and S2-glass fibers reinforced polycarbonate (Lexan 181, GE Plastics, Pittsfield, USA) prepregs were provided by ADM, a.s. (Brno, Czech Republic). Starch oil treated E-glass fibers (Vertex, CZ) with an average filament diameter of 12