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Mechanical performance of hybrid thermoset composites: Effects of matrix and reinforcement hybridization

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A B S T R A C T

Hybrid epoxy/vinyl ester (EP/VE) and epoxy/unsaturated polyester (EP/UP) resins were used as matrices to prepare unidirectional carbon fibre (CF) and carbon/glass fibre (CF/GF) reinforced composites targeting toughness of improvement. Hybrid resins were produced simultaneously (one-pot) and sequentially. (Thermo)mechanical properties of hybrid resins were determined in surface hardness, three-point bending, dynamic mechanical analysis (DMA), and differential scanning calorimetry (DSC) tests. Hybrid matrix composites with CF and CF/GF hybrid reinforcements were characterized with quasi-static mechanical (three-point bending) tests performed in 0° (longitudinal) and 90° (transverse) directions. In addition, flexural fatigue tests were run on UD composites. Interlaminar properties were deduced from in-plane shear strength (IPSS) test and fractographic inspection in a scanning electron microscope. The EP/VE hybrid resin exhibited improved energy absorption compared to neat constituent resins in contrast to EP/UP. Using hybrid resins as matrix highly improved the toughness and durability of the corresponding composites. Improved energy absorption was attributed to the phase structure of the hybrid resins, which also positively affected the IPSS.

1. Introduction

Fibre-reinforced polymer composites (FRPCs) have been greatly improved since they were first used in the middle of the 20th century. In the last few decades, FRPCs with enhanced properties have been developed to satisfy industrial demands [1]. One of the most important requirements besides high strength and modulus is toughness and mechanical durability. It is a challenge in the case of relatively brittle thermoset matrix FRPCs compared to ductile plastics, metals and alloys [2]. Hybrid resins as matrix material could be a feasible solution for this problem in cases where the relatively tough but limited strength thermoplastic polymer composites cannot be used. The special morphological structure of hybrid resins, widely called interpenetrating polymer network (IPN), increases the damping properties of composites, due to the entangled phases of the mixed resin. Because of phase conformation, this hybrid material can react differently to mechanical and thermo-mechanical loads compared to non-hybrid resins. The properties and possibilities of IPNs compared to neat resins have been investigated since the second half of the previous century [3–5]. The characterization of the formation of the network and thermo-mechanical proof of the phase structure have been published [5–8]. Numerous favourable attributes, for example enhanced damping properties [9–12], have been discovered and extensively analysed. Several kinds of polymer resin pairs were investigated, such as epoxy (EP) with polyurethanes (PU) [9,13–15], EP with unsaturated polyester (UP) [10,16], PU with UP resin [17,18], and EP with vinyl ester (VE) resin [11,19,20]. Hybrid resins have great damping abilities and some of them also showed synergistic mechanical properties. In addition, the hybridization of thermoset resins combines some of their favourable properties, for example good mechanical properties and the solvent-free processing of EP, excellent chemical resistance of VE, and the relatively affordable price of UP. Because of the increased damping abilities, hybrid resins could be a feasible matrix material of FRPCs. Although these hybrid materials composed of at least two resins have been in use for decades, their application as composite matrix is not common and has not been well published so far. FRPCs with a hybrid matrix have been investigated [21,22], but they focused mostly on the effect of surface treatment of the fibres and its impact on mechanical properties. Moreover, these studies used almost exclusively basalt

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and glass fibres (GF). Few papers have examined high-performance carbon fibre (CF) reinforced hybrid resin matrix composites and their morphological and mechanical properties [23]. In short, much could be done to improve the toughness and durability of FRPCs with a hybrid resin matrix. On the other hand, hybridization of the reinforcement is a keenly investigated subject in the case of thermoplastic [24,25] and thermoset matrix FRPCs [26–33]. These studies mainly focused on the enhancement of mechanical properties of thermoset matrix FRPCs but hybrid reinforcement also resulted in improved energy absorption and reduced delamination. Several types of reinforcement (mostly CF and GF) layers were used in different structural compositions. Combining hybrid matrix materials with hybrid reinforcement can result in even better properties. Therefore, this study investigates the effect of matrix and reinforcement hybridization in the case of thermoset FRPCs. The aim of this paper is also to investigate the toughening effect of EP/VE and EP/UP hybrids as matrix material. The EP/VE and EP/UP combinations were chosen because of the reduced styrene content compared to the neat UP and VE, and the reduced price compared to the initial EP. This study also examines the mechanical properties of hybrid resin matrix-hybrid CF/GF reinforced composites from a mechanical properties point of view.

2. Materials

A diglycidyl ether of bisphenol A type resin was used as EP with 188 g/epoxy equivalent and a density of 1.17 g/cm^3 (Ipox ER 1010, Ipox Chemicals, Hungary). Its hardener was isophorone diamine, with 43 g/hydroxy equivalent and ~660 mg KOH/g amine value (Ipox EH 2293, Ipox Chemicals, Hungary). An orthophtaleic acid-based resin with 39–42% styrene content was used as UP (DISTITRON-5119-ESX20ZQ, Polynt S.p.A., Argentina). A bisphenol-based resin with 35% styrene content was used as VE (AME 6000 T 35, Ashland S.p.A., Italy). The UP and VE resins were accelerated with 2 wt% methyl-ethyl ketone peroxide dissolved in diisobutyl phtalate (MEKP-LA-3, Peroxide Chem., South Africa).

Unidirectional (thin polyester yarn-stitched) carbon plies were used, with an areal weight of 309 g/m^2 CF in 0° and 10 g/m^2 GF in 90° directed rovings (PX35 UD300, Zoltek, Hungary). Carbon fibres of UD plies were sized for EP and VE and had a diameter of 8.3 ± 0.9 μm, a tensile strength of 2.48 ± 0.49 GPa and a tensile modulus of 133.5 ± 17.5 GPa. UD (thin glass filament-stitched) glass plies were used with an areal weight of 482 g/m^2 in 0° and 31 g/m^2 in 90° GF rovings (WR 482/31, Owens, Belgium). Fibres in the GF plies were sized for EP, UP and VE and had a diameter of 17.1 ± 2.9 μm, a tensile strength of 1.46 ± 0.70 GPa and a tensile modulus of 54.69 ± 9.07 GPa.

3. Experimental

3.1. Sample production and preparation

Based on previous investigations [7,8,19,23], 1:1 wt ratio mixed hybrid resins and two different procedures of mixing were used. One of them, called the simultaneous or one-pot method (henceforth referred to as method ‘A’), was described in detail in former publications [19,23]. Another method called sequential-like process (henceforth referred to as method ‘B’) was based on former studies [5]. The first step of the sequential-like method was the mixing of EP with its amine hardener for 2 min. Then the compound had a dwelling time of 30 min at 25 °C without stirring. After dwelling, the second resin (UP or VE) was added to the pre-reacted EP and stirred for 2 min. Then the curing agent of the second resin was added and stirred for 2 min again.

Resin samples were created with silicone moulds with a cross section of 4 mm × 10 mm. After moulding, specimens were kept in the mould at room temperature for 24 h. Composites were made by hand lay-up. 6 layers of unidirectional plies oriented in the same direction were used for CF and CF/GF reinforced composite specimens. The latter consisted of two outer CF “belts” (2 layers each) and an inner part of two GF layers. The benefits of this construction are low estimated decrease of stiffness and strength due to the CF “belts”, and expected energy absorption caused by the GF “core”. With this construction the coupling ability of hybrid resins at the CF/GF phase boundary can be observed (Fig. 1). Besides hybrid matrix composites, reference samples with identical reinforcement orientation and content were also prepared from plain EP, UP and VE resins. The designations of the materials, the resin content of the matrix, the layer build-up and the volume fraction of fibres are described in Table 1. The designation, mixing method and resin contents apply to resins and composites as well. The differences between the volumetric fibre content of CF and CF/GF reinforced composites may be an effect of the larger interfibre space between the glass (larger diameter) than the carbon (smaller diameter)

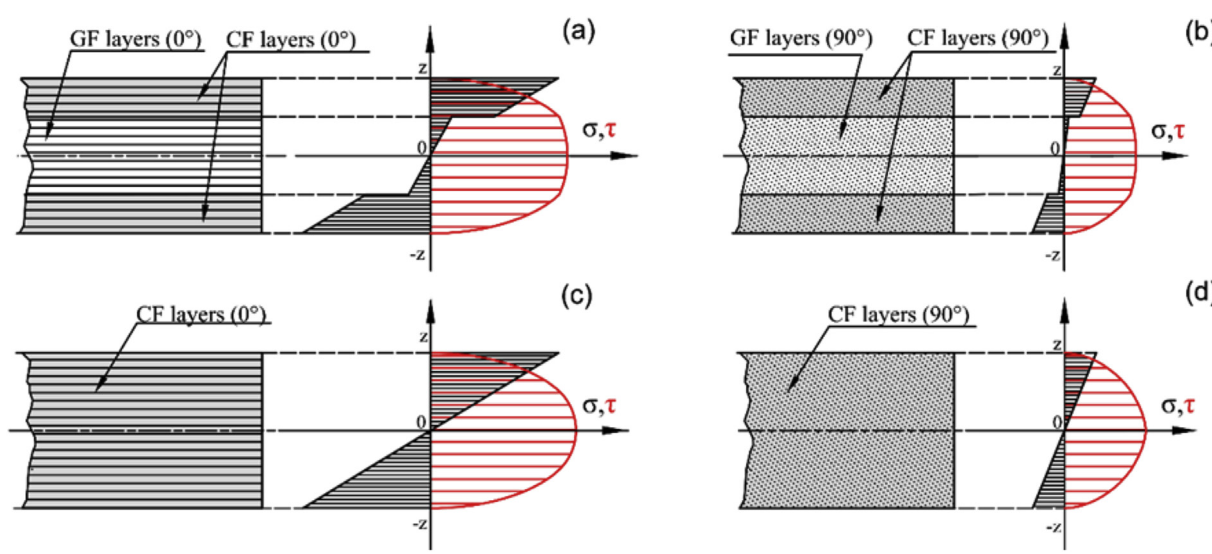

Fig. 1. Theoretical flexural stress distribution of 0° (a, b) and 90° (c, d) oriented CF and CF/GF reinforcement, (compression/tensile moduli presumed equal), based on [34].