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Quasi-static and fatigue performance of carbon fibre reinforced highly polymerized thermoplastic epoxy

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ABSTRACT

The experimental investigation aimed to understand the effect of the weight-average molecular weight of thermoplastic epoxy matrix on the quasi-static and fatigue properties of carbon fibre textile reinforced composites. The first part is dedicated to the assessment of the effect on the fibre and matrix adhesion by three scale measurements: single fibre and matrix (micro scale); single yarn and matrix (meso scale); laminate (macro scale). The positive effects at macro scale of the highly polymerised matrix are detailed in the second part considering quasi-static and fatigue tensile loadings. The composite with highly polymerised thermoplastic epoxy matrix showed an improvement of the tensile strength, a better capacity to uniformly distribute the stress/strain state and a considerable enhancement of the fatigue life.

1. Introduction

The development of thermoplastic resins has extensively increased the applications of carbon fibre reinforced thermoplastics (CFRTP) in aerospace, automotive and, as well as, consumer goods industries [1–5]. Their main advantage compared to thermoset counterpart are: increased toughness, better recyclability, and mainly the ability to deliver fast manufacturing processes [6,7]. To fully exploit the CFRTP potentials and to reduce costs, many researches and development programs were conducted, including physical and mechanical properties, as well as, short and long term behaviour (see e.g. Refs. [8,9]). However, available thermoplastic resins (TP) have higher melt viscosity than the thermosetting ones. As consequence, infusion of thermoplastics could not be an easy process leading to inappropriate impregnation of fibres. To overcome this issue, various impregnation techniques were studied. A solvent was proposed for reducing the viscosity of thermoplastics [10,11]. However, the solvent must be removed during manufacturing of the composite. Spraying TP powders as well as commingled yarns with TP fibres were developed to improve the impregnation of carbon fibres [12–15].

Recently, a new resin coupling the advantages of thermosetting and thermoplastic was developed [16,17]. It has the good workability of thermosetting and the post-formability and recyclability of thermoplastic. This thermoplastic epoxy (TP-EP) was adopted for CFRTP manufacturing [18]. The CFRTP produced with in-situ thermoplastic epoxy resin had a lot of attention [19], [20] being TP-EP without crosslinked structure. One of the main feature of a TP-EP is the weight-average molecular weight (Mw), which takes into account the molecular weight of a chain. It depends on the polymerization temperature and the polymerization time [21]. Mw is often used to estimate the physical properties of the resin and was connected to the mechanical properties of TP-EP [22,23].

The purpose of the present investigation is to give a contribution in understanding the effect of Mw of TP-EP matrix on the quasi-static and fatigue properties of CFRTP. The paper is subdivided in two parts. The first, preliminary, is devoted to the assessment of the influence of Mw on the fibre and matrix adhesion. For this purpose, three scale measurements were conducted: single fibre and matrix (micro scale); single yarn and matrix (meso scale); laminate (macro scale). Those were achieved by micro-droplet, matrix crack propagation thought a yarn interface and end notched flexure (ENF) tests. In the second part, the macroscopic effect of the enhanced adhesion of fibre and matrix was investigated by quasi-static and fatigue tensile loadings of the carbon textile reinforced thermoplastic epoxy with different MWs. Analyses were conducted with full field measurements by the digital image correlation technique (DIC) and local observations by Scanning Electron Microscopy (SEM) of the materials failure mode. The enhancement of the fibre and matrix adhesion observed at three scale levels was considered the main reason for the improvement of the quasi-static strength and for the considerable extension of the fatigue

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Table 1
Properties of thermoplastic epoxy resin and accelerator.

	DENATITE XNR6850A	accelerator XNH6850B
Chemical classification	Formulated epoxy resin	Aromatic phosphoric acid ester
Aspect	White paste	White powder
Viscosity at 25 °C	220 Pa s	Solid
Specific Gravity at 25 °C	1.17	1.10

life of the highly polymerized thermoplastic epoxy carbon composites.

2. Composites components and manufacturing features

2.1. Matrix and reinforcement

Plain weave carbon fibre fabric (Mitsubishi Rayon TR3110MS) was used as reinforcement (yarn TR30S 3L, linear density 1.79 g/cm^3, pick and end counts 12.5 per inch, areal weight 200 g/m^2, according to the producer). The textile reinforcement is selected to have an important influence of the matrix on the mechanical behaviour of the composites. It has a higher fibres and matrix adhesion surface than a unidirectional reinforcement.

Thermoplastic epoxy resin (DENATITE XNR 6850A) and the accelerator (XNH 6850B) were supplied by Nagase ChemteX Corporation, Japan [17]. The intrinsic properties of thermoplastic epoxy resin and accelerator are listed in Table 1 (according to the producer [17]) and T_g is approximately 100 °C.

2.2. Manufacturing of prepreg

Plain weave CFRTP prepreg preparation procedure had the following steps. The resin, 'XNR 6850A', was heated by using an electric oven at 120 °C. When the temperature of the resin reached 105 °C, the accelerator 'XNH 6850B' was added to the resin with stirring. After that, the plain weave carbon fabric was impregnated with the thermoplastic epoxy resin by hand lay-up. The Mw of prepreg was finally controlled by a predetermined time and temperature sequence [21].

2.3. Manufacturing of laminate

CFRTP prepregs, impregnated with the thermoplastic epoxy resin in the state of oligomer, were polymerized at a given temperature in an electric oven. The obtained prepreg was cut (245 × 245 mm) and dried at 50 °C for 12 h. CFRTP laminates were prepared by press moulding with 10 layers (tensile test) and 20 layers (ENF test) of dry prepregs by a heat-press device setting temperature of 175–195 °C and pressure in the range 6–12 MPa. The carbon fibre volume fraction of the CFRTP laminates was approximately 45%.

The adopted weight-average molecular weights of the thermoplastic epoxy matrix are listed in the synopsis of the experimental program given in Table 2.

Table 2
Weight-average molecular weight (Mw) of specimens used for each test ('k' means thousand).

Micro-droplet	26 k	34 k	53 k	70 k	90 k
Crack Propagation	35 k	73 k			
End Notched Flexure	14 k	54 k	63 k	108 k	
Quasi-static tensile	37 k	63 k	84 k		
Tensile-tensile fatigue	32 k	62-66 k	101-117 k		

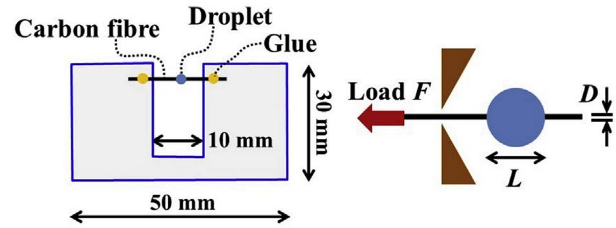

Fig. 1. Sketch of specimen and setup for micro-droplet test.

3. Experimental program

3.1. Measurement of weight-average molecular weight

The weight-average molecular weight of the thermoplastic epoxy matrix was measured for each batch by the gel permeation chromatography (GPC) adopting a CLASS-LC10 (Shimadzu Corporation) and a GPC column (Styragel HR4E, Styragel HR5E: waters). Tetrahydrofuran (THF) was used as solvent. The calibration curves were drawn based on the retention time and the Mw of standard polystyrene.

3.2. Micro-droplet tests

A scheme of the micro-droplet specimen and test setup is in Fig. 1. Both ends of a single carbon fibre were fixed on a sheet of paper using an epoxy-based adhesive. The droplet was attached to the carbon fibre by a soldering iron. The interfacial shear strength was determined with a pullout loading speed of 0.12 mm/min. The average strength was obtained using at least 20 specimens for each condition. The interfacial shear strength (τ) was evaluated by.

$$\tau = \frac{F}{\pi DL} \tag{1}$$

where τ is the interfacial shear strength; F is the pullout load; D is the fibre diameter; L is the adhesion length.

3.3. Crack development observation from a notch

Matrix and fibre debonding could be initiated by the development of transverse to the fibre cracks in the matrix grooving to the interface as the stress level in the fibre direction increases. As the applied load in the fibre direction increases, the cracks could grow along the fibre and matrix interface if the bond strength is weak. On the other hand, cracks could be arrested or slowed in propagation if the fibre and matrix has a high interface strength. Then, cracks could grow crossing the fibre, or could remain at the interface with increase of the load level, leading to the overloading and failure of fibre and consequently to the final failure of the composite.

To simulate this scenario and to assess the improvement of the yarn and thermoplastic epoxy matrix interface, increasing the Mw, the specimen depicted in Fig. 2 was designed. One yarn was embedded in the matrix by the following procedure. The yarn was extracted from the carbon textile. The yarn was placed on a release agent treated aluminium prismatic mould plate while a slight tension load was applied. Then, the TP-EP was poured into the mould where the yarn was set. The plate was kept in an electric oven for a predetermined time and temperature to get the specified Mw. After polymerization, TP-EP reinforced carbon yarn was removed from the aluminium mould. A centred pre-crack was introduced by a razor blade. During the tensile loading in the yarn direction of at least three specimens for each considered Mw, the Digital Image Correlation (DIC) technique [24] was adopted to estimate the evolution of the full field strain and to follow the propagation of the crack. For this purpose, the surface in the centre of the specimen for a length of about 30 mm was speckled with white and black acrylic paints (Fig. 2). A cold light source was used to