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Inclusion of a thermoplastic phase to improve impact and post-impact performances of carbon fibre reinforced thermosetting composites — A review

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ABSTRACT

Infusion processing methods have become a popular manufacturing alternative to the autoclave procedure to meet the increased demand for high-performance composites with shorter production times and lower cost. These processes are primarily limited to low viscosity, thermosetting matrices that are inherently brittle, and hence are susceptible to impact damage. It has been shown that introducing a thermoplastic modifier to create a "three-phase composite" can improve the ability of the laminate to resist damage formation and growth, and enhance a damaged laminate's structural performance. A comprehensive review is presented herein of the state-of-the-art on the incorporation of a thermoplastic phase into a fibre-reinforced thermosetting composite laminate to improve its damage resistance and tolerance properties when subjected to a low-energy impact. Several material properties govern the response of a laminate to an impact event, and for this reason, a discussion on the impact damage process and post-impact performance is also presented. Techniques from two main areas of toughening are considered — namely, bulk resin modification and interlaminar toughening. The improvements in laminate performance brought about by the thermoplastic additive are discussed, and each technique is assessed based on its suitability for inclusion in infusion manufacturing processes.

1. Introduction

Composite materials are being used increasingly in the manufacture of weight-critical structural components for the aircraft industry as they boast higher specific strength and stiffness than their metallic counterparts. Cost-effective composite structures can be manufactured using resin infusion (RI) techniques; RI manufacturing techniques include processes such as resin transfer moulding (RTM) and vacuum assisted resin transfer moulding (VARTM). A good design by RTM leads to the fabrication of three-dimensional near-net-shape complex parts, offering production of cost-effective structural parts in medium-volume quantities using low cost tooling [1]. The manufacture of larger structural components is made possible by VARTM, whereby one side of the tool is replaced by a flexible membrane, in order to reduce tooling costs. Due to processing considerations (i.e. viscosity, processing temperature, etc.), RI is almost exclusively limited to the use of thermoset resins that are inherently brittle and have relatively poor toughness, leading to the manufacture of composites with low damage tolerance that are susceptible to accidental impact damage — such as from runway debris, bird strikes or tool drops during maintenance. Impact damage causes serious deterioration in the structure's load-bearing capacity, brought about by matrix cracking, delaminations and fibre fracture. This can be a limiting factor in the structural applications of composite materials as the strength of composite structures drops significantly after impact because the loaded fibres are fractured and/or no longer adequately supported by the damaged matrix [2]. Delaminations are perhaps the most critical damage process in laminated composites as they are responsible for reducing the residual compressive strength [3,4]. Hence, the improvement and tailoring of composite properties for resistance to (and tolerance of) damage due to out-of-plane loading, such as impact events, have become an important research topic. The toughness of thermosetting resins can be improved by manufacturing a three-phase composite: fibre, matrix, and toughener [4]. The modification of a thermosetting composite with a thermoplastic toughening phase for improved resistance to impact damage has proven to be effective, and will be subsequently discussed. Modification via thermoplastic additives can be divided into two main categories: bulk resin modification and inter-/intralaminar toughening.

Much research has been conducted on bulk matrix toughening (Section 3.1) using core shell rubber (CSR), liquid rubber, polymer blends and hyper-branched polymers (HBP). Studies have shown that toughening the bulk matrix system with rubber can enhance the impact tolerance of the composite but can adversely affect other mechanical and thermal properties. Depending on compatibility with the matrix and the final morphology achieved, some thermoplastics (e.g. polyetherimide (PEI), polyethersulfone (PES)) when blended with epoxies do not sacrifice other properties, such as Glass Transition Temperature (Tg). However, the modification of the resin can potentially increase the viscosity, which is problematic for both prepreg and RI

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techniques. Electrospun thermoplastic nano-fibres located at the interlaminar regions of a composite have proven to be a highly effective toughening method [5–9], as they interact with and dissolve in the resin matrix during cure (thereby avoiding an increase in resin viscosity) and phase separate to form a toughened morphology, yielding large improvements in damage tolerance without adversely affecting other material properties. Hyperbranched polymers (HBP) also improve the toughness of the resin without significantly increasing the viscosity as they do not begin to phase separate (followed by an increase in molecular weight of the polymer during polymerisation, and hence viscosity) until the curing step starts. Inter-/intralaminar toughening (Section 3.2) is a broad term for a range of toughening techniques whereby the modifier is intended to reinforce the delamination-prone interlaminar region, e.g. co-mingled fibres, thermoplastic films, thermoplastic particles and non-woven fibre veils. Thermoplastic interlaminar particles have proven to be extremely effective for prepreg materials; the Boeing 787 fuselage, for example, is manufactured from a carbon/epoxy prepreg tape with thermoplastic particles located at the interlaminar regions [10]. The improvements achieved through the use of a thermoplastic toughener in a thermoset composite – specifically in carbon/epoxy composites – using both autoclave and out-of-autoclave manufacturing methods are well documented and will be discussed in the following sections. These improvements are primarily in Mode-I and -II strain energy release rate – or interlaminar fracture toughness (ILFT) – (G_{IC} and G_{IIC}), matrix fracture toughness (K_{IC}), interlaminar shear strength (ILSS), and impact damage resistance and tolerance. However, the effect of introducing a thermoplastic toughener into the composite material on other mechanical properties (such as stiffness), environmental resistance, processability and thermal properties (such as Tg) is important and needs to be considered.

The objective of this review is to compare the current toughening techniques for thermosetting carbon–fibre composites that use a thermoplastic modifier. They will be subsequently compared based on the suitability of each technique to RI processing. The approach aims to also give an understanding of damage progression through a composite material during a low-energy impact event and the material properties that are associated with this type of damage, i.e. G_{IC}, G_{IIC}, K_{IC}, ILSS, flexural and residual strength properties. The mechanism by which each toughening concept increases the damage resistance and damage tolerance of the composite will be discussed and published findings presented. Self-healing thermoplastics [11–17] were considered to be beyond the scope of this paper. This review focusses on carbon fibre-reinforced composites; the use of thermoplastic toughening additives to reinforce glass fibre composites is discussed in references [18–21]. It is well known that impact resistance can be improved through the use of different fabric architectures such as woven, spread-tow, and 3D woven fabrics. However, this is outside the scope of this review, which is concerned solely with the improvement in damage resistance and tolerance due to the thermoplastic toughening phase. Information on the use of these types of fabrics, and their effect on impact tolerance can be found in references [22–35].

2. Impact tolerance of composite materials

When assessing a candidate toughening concept for enhanced impact tolerance, it is important to consider impact damage mechanisms and how this damage affects the subsequent performance of the material. Greenhalgh and Hiley [36] consider impact tolerance to consist of two aspects: damage resistance (quantified by drop-weight impact testing) and damage tolerance (quantified by compression after impact (CAI) or open-hole compression (OHC) testing). The former describes the material's ability to sustain an impact with minimal damage, whilst the latter refers to the ability to retain structural performance once damaged. Therefore, to improve the impact tolerance of a composite material, an ideal toughening concept will resist the formation of damage due to impact and retain structural performance when damaged.

2.1. Impact performance — damage resistance

Investigating the process of impact damage formation is the first step to determining the key material parameters that govern the overall performance of the laminate when subjected to a low-energy impact. Tita et al. [37] proposed that impacts of varying energy can be divided into three distinct regions based on the resulting damage (Fig. 1): Region 1 (energy levels up to the impact damage threshold), Region 2 (energy levels that cause matrix cracks and delaminations only) and Region 3 (energy levels that cause fibre rupture and penetration). For the purpose of this paper, the authors consider the term “low-energy impact” to refer to an impact in which the damage imparted on the laminate does not extend to through-thickness penetration, i.e. Region 2. An overview of damage progression during a low-energy impact is provided herein. Details on mechanical investigations are available in references [36,38] and simulations of low-velocity impact damage progression were carried out by Tita et al. [37].

Composite laminates exhibit two distinct failure modes during a low-velocity impact: intraply failure in the form of matrix cracking and fibre/matrix interfacial failure, and interply failure (i.e. delaminations between plies). Fibre rupture is the dominant damage mechanism associated specifically with carbon and glass reinforced composites during high energy impacts due to their brittle nature [39]. Fig. 2 shows a schematic of the damage progression through a [0/90/0] laminate during a low-energy impact event. The resulting damage process can be divided into three main steps that occur in the following order: (1) fibre/matrix debonding and matrix cracking due to high transverse shear stress in the upper plies; (2) through-thickness bending crack due to high flexural stresses at the lower surface; and (3) delaminations due to cracks arrested and diverted along the interlaminar region.

Steps 1 and 2 involve intralaminar damage only, i.e. matrix cracking and fibre/matrix debonding. It can be seen that there are two types of matrix cracking formed during an impact: the first due to shear stress (Step 1) and the second due to bending stress (Step 2). This can be explained by considering the governing stresses for each step using a standard composite stress tensor. Matrix cracks initiate in the upper plies with the dominance of transverse shear stress, σ_{23}, and propagate downwards from the point of impact at an angle of approximately 45°. In the lower plies, the high flexural stresses (direct transverse stress, σ_{22}) induced by bending of the laminate during the impact create a through-thickness “bending-crack” [40]. Fibre/matrix debonding is influenced by the direct stress transverse to the fibre direction, σ_{22}, and shear stress along the fibre in the transverse and through-thickness planes, σ_{12} (Fig. 3). The five main mechanisms of intraply damage are shown in Fig. 4 (redrawn after Tita et al. [37]).

Barely Visible Impact Damage (BVID) is another characteristic of low-energy impacts, and is problematic to detect by standard in-service inspection methods as it consists of macroscopically small dents on a composite laminate surface that overlay significant internal damage [4]. As a result of this difficulty in damage detection, structures must have sufficient residual strength to resist failure at the design ultimate load to satisfy damage tolerance requirements. The small dents which constitute BVID are caused by debris lodging in matrix cracks formed during an impact event, preventing them from closing after the impact, thereby creating permanent indentations on the surface [41]. Hence, a key material property that influences the impact process is the fracture toughness (K_{IC}) of the matrix. Brittle matrices have little resistance to crack initiation and growth. By increasing K_{IC} of the matrix the resistance to crack growth will increase, thereby increasing the resistance of the laminate to delamination initiation caused by matrix cracking.

Step 3 of the damage process involves interply failure in the form of delaminations. Intraply damage (matrix cracks and fibre/matrix debonding) can initiate delaminations primarily due to a mismatch in properties between plies of varying fibre orientation. When a crack reaches the interface between two adjacent plies, the interlaminar