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Review article

Influencing the damage process and failure behaviour of polymer composites – A short review

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Abstract. Fibre-reinforced thermoset matrix polymer composites are gaining popularity in structural applications with strict quality requirements. However, their failure process, which is hard to predict and often catastrophic, mostly occurring at random locations, might be unfavourable in these areas. To ensure the further spread of composites and increase their reliability, their failure processes have to be influenced and controlled. This article gives an overview of different methods for influencing the damage process and failure behaviour of fibre-reinforced composite materials with thermoset polymer matrix. We describe the different methods, which can either control the failure in terms of location or mode or modify the failure behaviour, mostly by increasing toughness or achieving pseudo-ductile behaviour with a more gradual failure process. These methods can also simplify structural health monitoring, which has great importance in several applications of composite materials. The article focuses on the methods that enable the manipulation of the failure process by creating artificial damage or modifying the reinforcement, the matrix, the fibre-matrix interface, or the interlayer.

Keywords: *composite, damage, failure, pseudo-ductility, designed failure, artificial damage*

1. Introduction

Nowadays, polymer composites composed of fibre reinforcement and polymer matrix are rapidly gaining importance among structural materials and are now present in all areas of life. The popularity of polymer composites is mainly based on their outstanding mechanical properties – especially strength and stiffness – and their low density, which enable weight reduction compared to conventional materials. Their diversity is demonstrated by their applications ranging from everyday mass products, such as simple sports equipment and household appliances, to safety-critical engineering structures under significant loads, such as those used in construction, aerospace, aviation, water and land transport [1–5].

Although fibre-reinforced thermoset matrix polymer composites are widely used in load-bearing engineering structures, their further spread may be limited by their disadvantageous failure behaviour. Carbon fibre-reinforced composites are a good example, where, due to the low tensile elongation – generally under 2% strain-to-break – and complex microstructure of the reinforcing material, failure often occurs in a single step, catastrophically, and in random positions or often in several cross-sections simultaneously, which typically results in an explosive failure in unidirectional fibre orientations. Generally, failure of polymer composites with thermoset matrix is brittle after a linear elastic stress-strain response [6–9].

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The main problem is that the failure of composites is a very complex process, where different damage modes – fibre fracture, matrix cracking, fibre pull-out and debonding, delamination, *etc.* – can occur simultaneously depending on the loading conditions, which can lead to the failure of the structure. It is also problematic that damage in composites, especially delamination, is often difficult to detect and localise [10–13]. To further increase the use of composites and their reliability in high-performance applications and reduce the complexity of damage detection and structural health monitoring, which is important in these areas, it is essential to improve the controllability and predictability of their failure processes [14, 15].

There are several ways to influence the – micro- and macro-level – failure processes of composite materials, as illustrated in Figure 1. One method is to predefine the failure location. Suppose damage/failure can be isolated to a restricted zone predefined by the designer, and sensors are placed near this zone. In that case, it is possible to monitor the structural health of the composite part. In this way, failure can be predicted, or even prevented, if the damage is detected in time and the damaged component is taken out of service. Furthermore, localising the failure makes it possible to ensure that the damage in the composite part develops in an easily accessible location, thus ensuring repairability [16, 17].

The failure of composites can also be influenced by toughening methods, which result in an increased toughness and energy absorption capacity of the material. Furthermore, they might even ensure a more gradual release of energy accompanied by a gradual failure process. In this way, the damage process can be monitored to predict and prevent failure [9, 18]. However, in the case of fibre-reinforced thermoset matrix polymer composites, we cannot speak of ductile behaviour in the traditional sense, since the composite materials are incapable of significant plastic deformation. As an alternative, by modifying the composite appropriately, a material behaviour can be achieved, in which the stress-strain curve can be divided into two parts: a linearly elastic section and a nonlinear section similar to plastic deformation. This phenomenon is called pseudo-ductile behaviour, since the nonlinear relationship between stress and strain is not caused by plastic deformation but by a gradual damage process on the macroscopic level. These methods are used to modify the mode of failure, resulting in a more gradual failure process [9, 15, 17].

Another solution may be the combination of predefining the location of the failure and influencing the failure mode or even predetermining it as well, which could provide a designed failure in the composite material [17, 19] and a safety function of predefined fracture points, which can even help with the

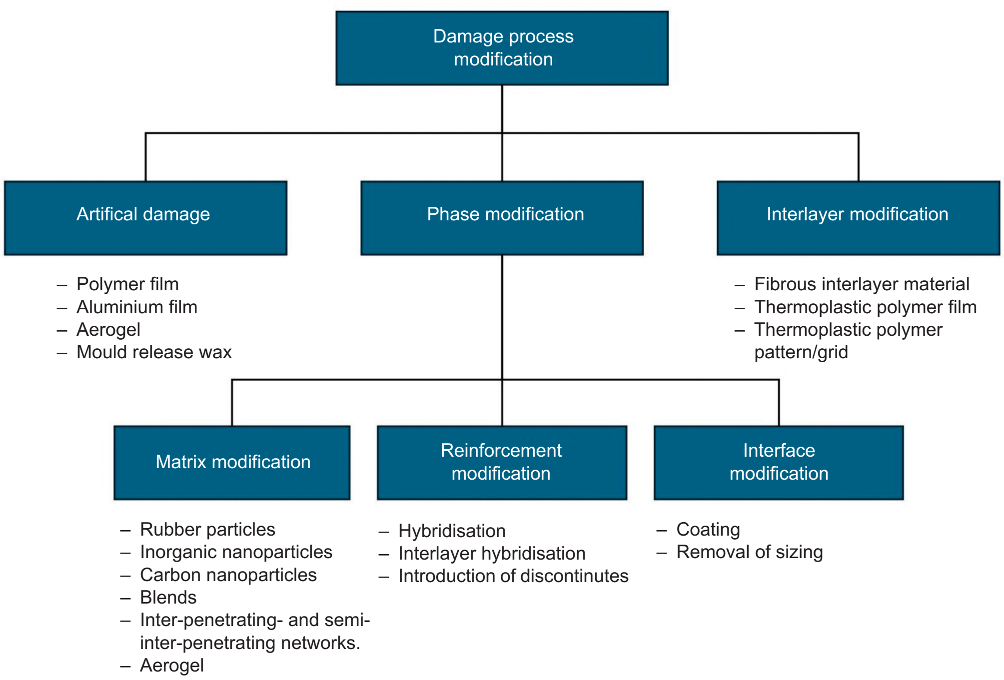

Figure 1. Classification of different methods used for influencing the damage processes and failure behaviour of composite materials.

recycling processes. An important question related to the application of these methods is the influence of the different processes used to modify the failure process on the key properties of the composite, particularly the mechanical properties. Furthermore, the different methods can have other non-negligible drawbacks, *e.g.*, increasing weight and manufacturing costs, adverse effects on thermal properties, decreasing durability, and manufacturing difficulties. It is essential that each advantage and disadvantage has to be taken into consideration when modifying the failure behaviour of a composite material.

2. Predefining failure location by artificial damage

The creation of artificial delaminations, where the interlaminar connection is locally interrupted by the application of a different interlayer material, which behaves similarly to real delamination, has been addressed in several research studies. These studies aimed to investigate the properties and failure behaviour of fibre-reinforced polymer matrix composites and to develop a more accurate material model for finite element simulations of damaged structures [16, 20–22]. However, in practical applications, introducing artificial damage can be a beneficial method for modifying the failure behaviour to enable the localisation of damage and failure to a predefined zone. On the one hand, this predefined failure location makes the failure of the composite safer, and on the other hand, placing sensors near this zone allows real-time tracking, and makes structural health monitoring of the composite part more effective [23].

Although damage localisation can be achieved by introducing artificial discontinuities in the composite, it is essential to note that the presence of the artificially induced damage has non-negligible negative effects on the mechanical properties of the composite, which is highly dependent on the loading conditions [13, 24].

Introducing polymeric or metallic films in the interlayer can create artificial delamination, thus localising the damage and ensuring failure appears at a predefined location. Mainly polytetrafluorethylene (PTFE, Teflon®), polyethylene terephthalate (PET), polyimide (PI) and aluminium films are applied for this purpose [21, 25–27].

In a research series [25, 26], PTFE and aluminium films were applied in the interlayer of carbon fibre-reinforced and glass-fibre reinforced epoxy matrix (CF/EP and GF/EP) composites to create artificial delaminations, to simulate real-life delamination damage. Compression test results show that applying PTFE or aluminium films to create artificial delamination can well approximate real-life damage. The results also indicate that the failure initiation point can be predefined through artificial delaminations. The presence of artificial delamination has a significant adverse effect on the mechanical properties under compression loading. Based on the compression tests carried out, it was found that the compressive strength decreases with increasing size of the artificial delamination. Furthermore, it has been shown that the closer the artificial delamination is to the mid-plane, the lower the load-bearing capacity of the specimen will be. The typical failure mode was buckling, and two different modes were observed, which were described as local and global buckling failure [28–30].

The failure mode can be influenced by the through-thickness position of the artificial delamination, where a location close to the mid-plane causes global buckling, while a greater distance from the mid-plane results in local buckling failure. For local failure, the buckling appeared only in the layers above the artificial delamination; in the other case, a classic global buckling failure was observed. Furthermore, in none of the arrangements was further propagation of the artificial delamination experienced before failure. This observation indicates that the failure process occurred in a localised manner [26, 28–30].

Polymer films such as PET and PTFE were applied in several studies to create well-defined artificial delaminations, and the delamination was detected successfully with non-destructive material testing (NDT) methods [16, 22, 31, 32]. The artificial delamination induced by the polymer films shows similar signals to the real ones during several NDT testing methods; however, there are some differences, which can make detecting real damage based on data collected on artificial damage challenging [16, 33]. Infrared thermography and acoustic emission testing identify artificial damage successfully [16, 27]. While applying the digital image correlation (DIC) method during low-strain loading as an NDT method, the information indicating the presence and propagation of delamination damage is provided by the high local strain values and the changes in the strain field observed during full-field strain measurements [34]. In the case of polymer films, the NDT-DIC method also enables the detection of artificial damage at low strain values in the non-destructive region [27]. However, the method is unsuitable for other artificially created discontinuities, such as mould release wax encapsulated in the composite, which does not cause a well-defined delamination – and a localised failure [22].

Another, more innovative method for creating artificial delaminations can be using aerogels. Silica aerogel is an ultralight, porous, gel-like material that contains 95–99% air by volume, resulting in an exceptionally low density. A real delamination can be described as an elongated crack or cavity containing air. Thus, the artificial delamination created by placing a small, thin piece of aerogel in the interlayer reproduces the real delamination better than previously developed solutions, which might also be an advantage while providing localised failure. The artificial damage made by aerogel can also be more easily and more accurately detected with NDT methods such as thermography and ultrasonic testing, and its signals are more similar to real damage than in the case of artificial delamination caused by polymer films [20].

Damage and failure localisation has a further advantage in facilitating the repair of the composite part. Thus, if the damage is localised to a well-accessible location and a small area, the part can be repaired more easily and quickly, even on-site, by patch or scarf repair [24].

The studies presented in this chapter show that by using various interlayer materials, artificial delamination can be created that behaves similarly to real damage and can provide a localised failure or serve as a starting point for further delamination, thereby localising the damage to some extent. The disadvantage of these methods can be their influence on the mechanical properties, which greatly depend on the loading conditions. However, there is a possibility of improving interlaminar properties, modifying the stress-strain response, or even create pseudo-ductile behaviour through artificial defects.

Herráez *et al.* [35] investigated the possibility of introducing PTFE films as small interlaminar defects to improve the fracture toughness of autoclave-cured unidirectional (UD) carbon/epoxy prepreg-based composites using double cantilever beam (DCB) and mixed-mode bending (MMB) tests. They managed to enhance the Mode I fracture toughness by up to +430% and the Mixed-Mode fracture toughness by up to 115% by placing 1–3 artificial defects into the interlayer. The mechanism can be explained by the simultaneous, stable propagation of more than one crack along different interfaces, which results in the increase of dissipating fracture energy. Furthermore, the artificial interlaminar defects can enhance fracture toughness by increasing the local mode variety at the crack and activating other toughening mechanisms such as fibre bridging. However, as a disadvantage of the method, the introduction of localised interlaminar defects significantly reduces the bending stiffness. The experimental tests were supported by finite element analysis using cohesive zones, demonstrating similar results.

Melaibari *et al.* [36] created bio-inspired laminates with embedded defects, containing PTFE film-induced delaminations with different sizes at different through-thickness positions, and investigated their behaviour with three-point bending tests. With proper placement and size of the artificial delamination, the composites demonstrated a progressive damage mode with pseudo-ductile response, which can be explained by a predefined crack initiation point and altered crack propagation, where the propagation of delamination, matrix cracking and the breakage of fibres reaches the different interlayers and plies gradually. Regarding mechanical properties, most of the laminates show properties similar to those of the baseline laminates. Furthermore, the investigated properties can be improved depending on the delamination size and position. Laminates with a 10 mm delamination diameter located in the nearest interlayer to the indenter demonstrated the best results with 11.9, 208 and 288.1% enhancement in flexural strength, failure strain and energy absorption capacity, respectively.

3. Modification of the reinforcement and/or matrix material

The required failure behaviour, in the case of most research studies, pseudo-ductile behaviour, can be reached through the modification of one phase of the composite, the matrix, the reinforcement material or the interface.

3.1. Modification of the matrix material

In the case of thermoset matrices, the cross-linked structure generally results in a brittle material with low impact and fatigue resistance and poor fracture toughness, in which preexisting cracks can easily propagate. Several methods can hinder crack propagation, resulting in increased toughness and modified failure behaviour of the matrix material and, thereby, of the composite [37, 38].

One way to increase the toughness of polymer composites can be to introduce a second, well-dispersed phase. A common method is the application of rubber particles, which can effectively increase the toughness of the matrix and, thus, the composite material [39–43]. The rubber and resin phases' compatibility fundamentally determines the connection quality between the two phases, which can be improved by the surface treatment of the additives [44]. The toughening mechanism is achieved due to the well-dispersed rubber particles forming a second phase, thereby creating elastic regions inside the matrix material, where the deformation and fracture of the particles and the plastic void formation result in the modified behaviour: when the propagating crack reaches such a highly elastic zone, it can be pinned [45, 46].

Wang *et al.* [45] applied carboxyl-terminated butadiene-*co*-acrylonitrile (CTBN) particles in different contents to increase the toughness of a bisphenol F diglycidyl ether (DGEBF) based epoxy resin using a pre-reaction method to ensure the proper chemical bonding on the interface between CTBN and EP. The single edge notched bending (SENB) tests determined the Mode I fracture toughness. The addition of 15 phr CTBN demonstrated the best results, with 104.1 and 5.2% improvement in fracture toughness and tensile strength, respectively.

Relevant studies show that the matrix's glass transition temperature (T_g) can also be significantly reduced by the added rubber particles, leading to the system's plasticity. The results of several studies show that adding rubber particles to the epoxy matrix can significantly increase the energy absorption capability and impact resistance of the material, and it may also help improve the crack propagation resistance of the composite. Another effect of adding rubber particles is that due to their low modulus and strength, the Young's modulus and yield strength of the epoxy are also reduced [40, 43, 47].

Using inorganic nanoparticles with higher stiffness can also improve the fracture properties of epoxies in an opposite process to rubber-like particles. These high-modulus, rigid particles also induce an increase in the strength and modulus of the epoxy. In the case of nanoparticles, silica, glass or alumina particles are common [40, 42].

According to other researchers, the toughening mechanism in epoxy composites modified with silica nanoparticles is similar to that in epoxies toughened with rubber particles. A localised ductile shear zone is formed under the effect of concentrated stress acting around the silica nanoparticles, which results in behaviour with increased toughness [43, 48]. In general, using silica nanoparticles for toughening is a less efficient method than using rubber particles [40, 41]. Silicate nanoparticles have been applied in combination with rubber particles to modify the failure behaviour of epoxy as well, resulting in increased fracture toughness of the material [39, 43].

There were also several attempts to modify the failure process of the composites with the addition of graphene [49–52], graphite nanoplatelets [53, 54], carbon nanofibres [55] or carbon nanotubes [56, 57]. The use of these particles in polymers is becoming increasingly common due to their high specific surface area, resulting in outstanding electrical, thermal and mechanical properties, which enable the development of multifunctional composites [58–61]. Their use with various polymers, including epoxy, has become widespread in recent years. The addition of nanoparticles can contribute to an increased Mode I fracture toughness of the material by slowing down crack propagation due to an altered crack path with an increased length [62, 63] (Figure 2). Tareq *et al.* [63] experienced a 40% increase in Mode I fracture toughness – conducted by DCB test – by adding 0.1 wt% graphite nanoplatelets (GnP) – mixed to the A-component of the epoxy resin system – to the matrix of the composite.

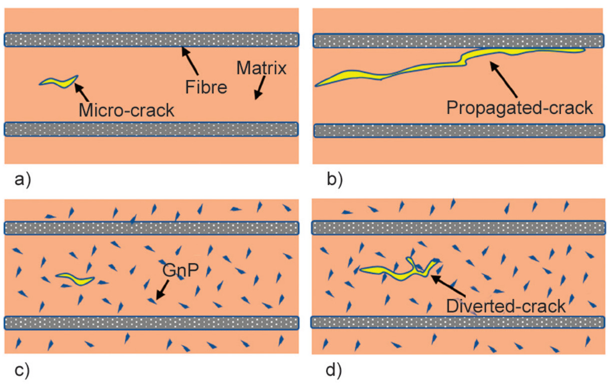

Figure 2. Altered crack propagation in carbon-fibre reinforced composites containing graphene nanoplatelets (c), (d) compared to reference composites without graphene (a), (b) [63] (reprinted with permission of Wiley).

Chandrasekaran *et al.* [64] investigated the effect of three different types of nanofillers on the fracture toughness and the damage mechanism of epoxy-based nanocomposites. They measured the effect of thermally reduced graphene oxide (TRGO), GnP and multi-walled carbon nanotubes (MWCNT) and studied their effect on mechanical properties. The fracture toughness was measured as a function of the filler content in weight percentage with the end-notched flexure (ENF) test. They experienced a significant growth in fracture toughness by adding up to 2 wt% nanoparticles to the system. However, in the case of TRGO and MWCNT, they could not achieve a filler content higher than 0.5 wt% based on processing limitations due to the high viscosity of the nanocomposite suspension. It can be noted that graphene oxide had the highest toughening effect, which was 40% at 0.5 wt%.

Polymer blends can provide another solution. Blends can be produced to increase the toughness of the composite by mixing epoxy resin with a thermoplastic polymer. The thermoplastic polymer is mixed into the matrix material as fibres or granulates and then melted during high-temperature curing [65, 66].

Blends can successfully toughen the matrix material, thereby increasing fracture toughness and, in several cases, improving fatigue resistance, without significantly reducing strength and elastic modulus. Experimental observations indicated that ductile thermoplastic particles contribute to the toughening mechanisms in thermoplastic/epoxy blends in two key ways: plastic deformation of the material surrounding the macroscopic crack tip and forming particle bridges within the crack path [65, 67].

Some polymers used for blend formation are polyphenylene oxide (PPO) [68], polysulfone (PSF) [69], polyethyleneimine (PEI) [70], ethylene methyl methacrylate (EMMA) [71], ethyl phenyl acetate (EPA) [72], polystyrene (PS) [73], polyether ketone (PEK) [74], polyether imide (PEI) [75], polybutylene terephthalate (PBT) [75] and polyamide-12 (PA12) [76]. Several copolymers – mostly block copolymers or even random ones–have also been used for toughening epoxy matrix materials [77–80]. Furthermore, even blends such as the widespread PC/ABS blend might be applied in this area, resulting in a modified crack formation and propagation and a less brittle matrix behaviour, thereby increasing fracture toughness and impact resistance [81–83].

The effectiveness of the blends in modifying the failure process can be explained by two mechanisms: the addition of a second phase with higher toughness and the altered crack propagation [65]. The formation of epoxy/thermoplastic blends has a further advantage. Such systems can also have a self-healing function since the thermoplastic polymer can be melted again and fill the cracks of the damaged material to obtain a functional composite with reduced load-bearing capacity but still largely structurally intact [72, 84, 85].

Inter-penetrating networks (IPN) can also be formed to modify the matrix material. With IPN systems, it is possible to create hybrid matrices that display the favourable properties of the different matrix materials simultaneously; thereby, the toughness of the original matrix material can be increased. An IPN network is a 3-dimensionally structured matrix material system with two or more phases (Figure 3). IPN networks are characterised by the interlocking of two phases, which are continuous and, therefore, cannot be separated. There is no primary, covalent bond between the phases in the system; there are only secondary bonds [86–88].

Numerous research studies were conducted on the formation of epoxy-vinyl ester (EP/VE) IPN systems as a toughening method of epoxy matrices. The studies were successful in creating IPN networks with different ratios of the two resins, thereby significantly increasing the fracture toughness of the epoxy resin. The formation and the stability of the IPN network, thus the increment of the fracture toughness, depends on the mixing ratio of the two materials [90–92]. Epoxy can form an IPN structure with polyurethanes (PUR) as well, where the PU chains can have a high degree of mobility, which might result in increased energy dissipation during fracture [87, 93–95].

Thermoset materials with cross-linked structures can form a so-called semi-IPN with thermoplastic polymers, similar to IPN systems. They can provide a

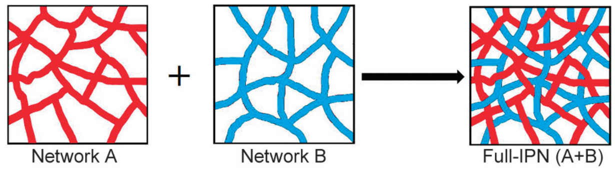

Figure 3. Schematic drawing of an interpenetrating networks consisting of two phases. Reprinted with permission from [89] (reprinted with permission of ACS).

successful solution for modifying the matrix material and the composite. Materials such as polycaprolactone (PCL) [96–99], polyethersulfone (PESU) [100, 101], polyimide (PI) [102] and further polymers [103, 104] and copolymers [105, 106] have been applied recently to increase the toughness of the epoxy matrix, thus the composite material. In the case of adding the thermoplastic material to the epoxy matrix during the curing process, phase separation occurs, and depending on the proportion of the thermoplastic and the resin and the parameters of the curing process, a semi-IPN might be formed. The resulting structure leads to increased fracture toughness and energy-damping capacity and might cause an increment in further properties such as tensile strength and impact strength as well [105]. Epoxy/thermoplastic semi-IPN structures are also used for self-healing [99, 107].

A novel method for toughening epoxy with the creation of a semi-IPN structure can be the application of thermoplastic epoxy (TPE). Due to their similar chemical structures, TPE exhibits excellent compatibility with bisphenol A diglycidyl ether (DGEBA) based thermosets. The introduction of TPE can improve several mechanical properties, such as tensile strength, flexure strength, impact strength and fracture toughness of the matrix material, which is associated with the formation of the semi-IPN structure of TPE and DGEBA-based epoxy. It is important to highlight that no phase separation occurs as the TPE content increases. This can be attributed to the similarity in chain structure and the strong hydroxyl interactions between the TPE and epoxy thermosets [108].

The TPE content highly influences the mechanical properties. Cheng *et al.* [108] added TPE to a DGEBA-based epoxy resin system in different amounts of up to 20 phr (parts per hundred). They experienced a slight – lower than 10% in each case compared to the reference composite – decrease of tensile and flexural moduli with increasing TPE content. However, several mechanical properties show maximum increase at different TPE contents – tensile strength, flexural strength and fracture toughness at 15 phr, while impact strength at 10 phr. These phenomena can be explained by different simultaneous effects. The formation of a semi-IPN structure and the strong intermolecular force between hydroxyl groups of TPE and hydroxyl groups formed during the epoxy curing process contribute to the improvement of strength and ductility. However, a significant part of hydroxyl groups responsible for curing reaction are bonded in intermolecular connections, which results in the reduction of cross-link density of the thermoset epoxy resin, which has a negative effect on several investigated mechanical properties [109].

The matrix material and, thereby, the fibre/matrix interface of continuous glass fibre/epoxy composites can be modified by nanostructured silica-based aerogels, resulting in a three-phase hierarchical structure. The aerogel's compatibility with silicate fibres enhances the interlaminar properties and compressive strength by up to 27% without reducing stiffness, while adding minimal weight (~1.3%). The continuous aerogel network ensures efficient load transfer and fibre support, hindering micro-buckling of the fibres [110].

3.2. Modification of the reinforcement material or the fibre-matrix interface

A modified and controlled failure of the composite can be provided by the reinforcement material as well, which is often achieved by hybridisation, where the advantageous properties of two or more reinforcement materials might be combined, *e.g.*, carbon fibre, combined with a reinforcing material, which has a higher deformability, such as aramid [111–113], glass [114] or natural plant fibres [115, 116]. Combining glass fibres with plant fibres is also common [117]. The hybridisation can improve several mechanical properties *e.g.* deformability, energy-damping capacity, fatigue resistance or fracture toughness of the composite. However, as a disadvantage of this method, a significant decrease might be experienced in some mechanical – mainly strength and modulus – and thermal properties compared to non-hybrid composites.

Interlayer or sandwich hybrid composites were developed mainly using prepregs. In these composites, usually, plies reinforced by a low-strain material, mainly carbon fibre, are combined with plies reinforced by a high-strain material, mainly glass fibre, in order to achieve pseudo-ductility. The pseudo-ductile behaviour is based on the different modulus of the two materials and the proper geometrical design – especially in terms of thickness – of the different layers. Pseudo-ductile behaviour is demonstrated by a plateau in the stress-strain response caused by the fragmentation of the thin carbon fibre reinforced layer and the stable delamination between the carbon fibre and glass fibre reinforced layers [118, 119].

Interlayer hybridisation can ensure the pseudo-ductile behaviour, thus a gradual failure under several loading conditions with different reinforcement orientations, *e.g.*, uni- [120], bi- [121], multidirectional and quasi-isotropic [122] as well. The damage initiation in interlayer hybrid composites can be made visually detectable without a catastrophic failure occurring during the plateau. This enables the use of these hybrid composites as structural health monitoring sensors [121, 123]. Interlayer hybrid composites can be applied similarly to achieve pseudo-ductile behaviour in the case of impact damage as well [124]. The interlayer hybridisation method for manufacturing pseudo-ductile composites was successful with the application of only carbon fibres as well, where two types of the same reinforcement fibre with differing modulus serving as low-strain and high-strain material, thereby meeting the design requirement on the material side. [125, 126].

Hierarchical hybrids with pseudo-ductile behaviour can be manufactured by combining continuous high-elongation fibres with intermingled hybrids made from highly aligned discontinuous fibres with lower elongation, where the discontinuous fibres are mostly aligned with a flow-induced alignment technique ensuring the desired fibre orientation and uniform fibre dispersion. Yu *et al.* [127] applied continuous S-glass (cSG) layers, and discontinuous high modulus carbon (HMC) and high strength carbon (HSC) or E-glass (EG) fibres aligned with high performance discontinuous fibres (HiPerDiF) method [128], which is based on the use of a low-viscosity medium, *i.e.* water instead of high-viscosity media during the alignment process, in the intermingled hybrid layers, resulting in pseudo-ductility. The damage mechanisms leading to the failure of the composites were highly influenced by the behaviour – linear or non-linear stress-strain response – of the intermingled hybrid layer.

Another method of modifying failure behaviour is to cut the fibre reinforcement causing ply-level discontinuities. By local cutting of the fibres in different patterns, the system is able to undergo higher deformation, because the fibre breaks occur only at a higher strain level. The failure process is dominated by matrix cracking and delamination, which results in a more gradual failure. Depending on the pattern, even other mechanical properties, such as tensile strength, might be improved [129].

Bullegas *et al.* [130] introduced patterns of micro-cuts into the microstructure of cross-ply and quasi-isotropic (QI) thin-ply carbon fibre-reinforced polymer (CFRP) composite laminates, aiming to improve their damage resistance under various loading conditions, based on a novel finite fracture mechanics model. Their method resulted in a significant improvement in terms of the notched strength of the laminate and the translaminar work of fracture during compact tension tests for CP laminates and QI laminates, and an increase of the total energy dissipated during quasi-static indentation tests on QI laminates. It has to be emphasised that the crack path and the damage modes were greatly influenced by the introduced micro-cuts.

Czél *et al.* [131] developed pseudo-ductile prepreg-based UD carbon/epoxy composites with overlapped ply-level discontinuities. The composite specimens were tested under quasi-static tensile loading. The mechanical response and the failure process were highly influenced by the geometry of the overlaps. The long overlap specimens demonstrated a non-linear stress-strain response based on the progressive interlaminar damage. Meanwhile, short overlap specimens showed an interfacial strength-driven failure with no interlaminar cracking. In terms of mechanical properties, the short overlap specimens failed at low stresses – 322 MPa – compared to the tensile strength of the continuous reference composite – 2724 MPa – with an almost 40% decrease in initial modulus. In case of long overlaps, the introduction of the ply-level discontinuities led to a less than 10% decrease in initial tensile modulus while retaining a reasonably high 1009.6 MPa tensile strength.

The introduction of ply-level cuts can contribute to the creation of a so-called bio-inspired 'brick-and-mortar' structure or hierarchical 'brick-and-mortar structure' inspired by the structure of nacre in nature (Figure 4). The application of this design method leads to a non-linear stress-strain response due to progressive interlaminar damage and enables control over the initiation and propagation of cracks, helping to minimize the typical catastrophic failure, and can contribute to the formation of a gradual, pseudo-ductile failure mode [132, 133]. The method might also be applicable at industrial levels, *e.g.*, it might be integrated into automated fibre placement process by applying predefined or even different fibre lengths [134].

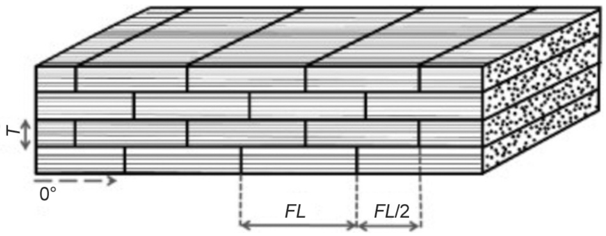

Figure 4. Schematic of a 'brick and mortar' structure in case of UD composutes (*T* – brick thickness, *FL* – fibre length, *FL/*2 overlap length) [134] (figure licensed under the Creative Commons Attribution 4.0 International License (CC BY 4.0)).

A further concept of enhancing the toughness is to modify the fibre-matrix adhesion. Interfacial adhesion is inevitable in the load transfer in the composite. The choice of sizing can influence the load transfer and the performance of the composite. With good quality adhesion, a matrix crack propagates to the interface and then to the fibres, causing localised fibre fracture, resulting in brittle failure [135].

If interfacial adhesion is deliberately weakened, the crack will follow the interface, as less energy is required for the crack to propagate along the interface than through the fibre. Thus, the crack travels a longer distance, resulting in a more gradual failure behaviour, but at the same time, as a compromise, the tensile strength is naturally reduced. By controlling the interfacial adhesion, an optimum can be achieved where a desired combination of high strength and pseudo-ductile behaviour can be ensured [136, 137].

Several studies show that the sizing of the reinforcement fibres can determine the fibre-matrix interfacial adhesion, thereby modifying the failure behaviour as well. The manipulation of interfacial adhesion for achieving pseudo-ductile behaviour was investigated on carbon fibres by coating the surface of carbon fibres with polyurethane (PUR), where the coating successfully prevented the catastrophic failure of the fibres by promoting crack propagation along the interface, *i.e.*, debonding and delamination, thereby increasing fracture strain of the composite as well [136].

A similar effect of weakened adhesion can be created by fibres with alternating adhesion strengths by coating the fibres with an adhesion-weakening material. Atkins [138, 139] coated boron fibres periodically in smaller regions with PUR. The application of the modified reinforcement material in polymer composites resulted in a significant ~400% improvement in fracture toughness with an acceptable less than 10% reduction in tensile strength.

In the case of interpenetrating networks, the fibre-matrix interfacial adhesion can show significant differences depending on which of the two phases of the matrix is in connection with the fibre. These local deviations might influence the crack propagation as well [92].

It has to be emphasised that the concept of creating pseudo-ductile material behaviour by modifying the fibre-matrix interface may negatively affect the load-bearing capacity of the fibres and, thus, the composite structure; therefore, such methods require thorough design and comprehensive characterization of the material structure and properties of the material.

However, the removal of the sizing can be a well-designed interface engineering technique as well, which might be used for manipulating crack propagation and modifying the failure of the polymer composite. Unlike conventional methods, which remove the sizing entirely, such as burning or using solvents, the modification of the interface is possible in a predefined pattern by using a CO_2 LASER. The pattern consisting of weakened adhesion zones is able to alter crack propagation by forming local delaminations. In the case of applying this technique only locally, restricted to a small region, the method can also provide a solution for the creation of localised damage [140].

4. Modification of the failure behaviour with interlayer methods

The interlaminar behaviour and the corresponding interlaminar properties, *e.g.*, interlaminar shear strength (ILSS), fracture toughness, fundamentally determine the damage and failure processes of a composite. In addition, the presence and propagation of manufacturing-induced flaws/defects in the composite, such as matrix cracks or voids, and the effects of in-service loads, vibrations and impacts can cause delamination, which significantly reduces the strength and the stiffness of the composite and can lead to failure [141, 142].

For all these reasons, it is important to manipulate the interlaminar behaviour of the composite, to increase its resistance to interlaminar crack propagation and thus avoid sudden catastrophic failure. A solution to this may be the introduction of interlayer material, which provides the composite with greater deformability and energy absorption capacity, and typically a gradual failure, based on a modified failure mechanism and crack path. Several types of interlayer materials are widely used [143].

4.1. Fibrous interlayers

One method is similar to the hybridisation of the reinforcement material. In this case, interleaving is realised by the application of a nanofibrous interlayer, a microfibrous interlayer or a multiscale interlayer material consisting of combined nano- and microfibres. The interleaving is responsible for the alteration of damage mechanisms and crack propagation, thereby increasing the length of the crack path and improving the interlaminar properties of the composite.

Natural fibres are commonly applied in this area. Interleaving UD flax/epoxy composites with randomly oriented chopped flax yarns can improve the Mode I fracture toughness of the composite, which is explained by modified damage mechanisms, thereby increasing energy dissipation during delamination. It has to be emphasised that the process is greatly influenced by the length of the chopped yarns. [144].

Zhang *et al.* [145] investigated the application of a multiscale fibrous interlayer consisting of chopped flax fibres and waterborne epoxy-treated cellulose nanofibres. The application of the interlayer has led to a significant 122% improvement in terms of Mode II fracture toughness of the composite material – evaluated by ENF tests – compared to the original CFRP. This enhancement can be explained by the synergistic effects of the flax fibres and the cellulose nanofibres. Flax fibres hindered the agglomeration of the nanofibres, while cellulose nanofibres improved the flax/epoxy interfacial adhesion. These phenomena contributed to altered damage mechanisms and resulted in the occurrence of fibre bridging, fibre pullout and fibre fibrillation, thereby increasing the delamination resistance.

Furthermore, carbon nanofibres can be applied to influence the interlaminar behaviour. In the study of Arai *et al.* [146] the application of vapor-grown carbon nanofibre interlayers has demonstrated great potential in improving the fracture toughness of prepreg-based, autoclave-cured carbon fibre/epoxy composites with up to 50% increase in Mode I and up to 200% Mode II fracture toughness in comparison to the baseline laminates. In addition, both Mode I and Mode II fracture toughness have shown a significant dependence on the interlayer thickness.

Aramid, as a tough polymer fibre, widespread for toughening of CFRP through hybridisation, is a common interlayer material as well. Cheng *et al.* [147] investigated the effect of ultra-thin interlays with nonwoven aramid pulp micro-/nanofibres under uniaxial compression loading. Compression strength and compression strength after impact (CAI) significantly increased as a result of interleaving. An optimal areal density – which is related to interlay thickness – of the interlayer material was investigated in the research as well in the case of 0, 2, 4, 6 and 8 g/m^2 areal densities. 6 g/m^2 areal density demonstrated the most remarkable improvement of the investigated mechanical properties. Furthermore, the mode of failure was influenced as well, leading to a classical shear failure with few delaminations instead of internal cracking and delamination under the applied loading conditions, as shown in Figure 5. The phenomena can be explained by the aramid fibres filling resin-rich areas and creating fibre bridges at ply interfaces, effectively improving delamination resistance. Interleaving with aramid pulps can also be effective in increasing the impact resistance of the composite [148].

Several other polymer nanofibres proved to be effective as interlayer materials in toughening composites. Molnár *et al.* [149] studied the effect of electrospun polyacrylonitrile nanofibrous interleaves,

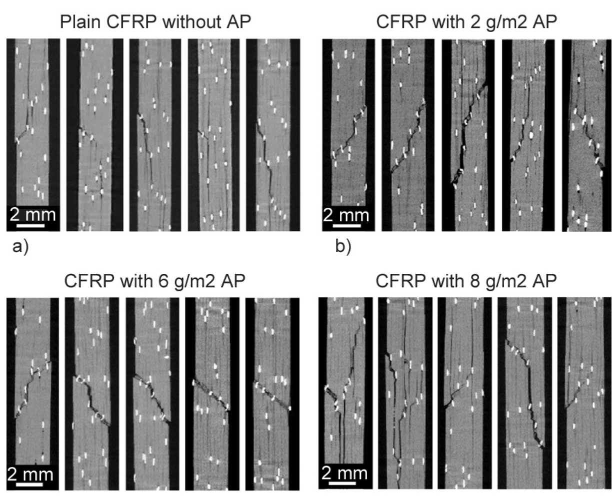

Figure 5. X-ray micro-computed tomography scans of failed specimens with different areal densities of the interlayer under uniaxial compression loading. a) Plain CFRP without AP, b) CFRP with 2 g/m^2 AP, c) CFRP with 6 g/m^2 AP and d) CFRP with 8 g/m^2 AP [147] (figure licensed under the Creative Commons Attribution 4.0 International License (CC BY-NC-ND 4.0)).

which were produced directly on the surface of the reinforcement material, on the flexural, interlaminar and impact behaviour of carbon fibre/epoxy composites with different reinforcement types (UD, woven). They experienced significant improvements in the investigated properties based on the energy absorption behaviour of the interlayer and its ability to contribute to the load transfer between carbon fibres.

Some thermoplastic polymers, which are widely used for the toughening of epoxy through blending or even the creation of a semi-IPN structure, can be applied in the form of interlayers as well. Cheng *et al.* [150] applied melt-spun, uniformly aligned PES fibre webs as interlayer material in carbon fibre/epoxy composites. Polyethersulfone (PES) nanofibre interleaves were wetted by the epoxy resin, leading to their dissolution and in-situ phase separation upon a concentration gradient around the reinforcement fibres, thereby increasing toughness in the interlayer and improving Mode I and Mode II fracture toughness, ILSS and CAI strength compared to the reference composite up to 103, 68.8, 18 and 43% respectively. Even tensile strength demonstrated a slight – 7% maximum – increase with the use of the PES interlay, although the tensile modulus decreased up to 20%. The decrement of the modulus can be explained with the application of the PES interlay reducing the fibre volume fraction of the composite.

PCL nanofibrous interleaves were investigated in several research works. Relevant studies show that the use of electrospun PCL nanofibrous interlayers leads to a significant increase in Mode I fracture toughness of multiaxial carbon fibre/epoxy [151] and glass fibre mat/epoxy [152] composites. In the case of UD glass/epoxy prepreg-based composites [153], besides Mode I increment, a significant improvement of Mode II fracture toughness by a similar amount – 25 and 24% – is reported as well. Meanwhile, the results of the research works indicate that the use of the PCL interlayer does not significantly influence the tensile, shear and dynamic mechanical properties. The effects of the interlayer are influenced by the concentration of PCL solution used for electrospinning, which phenomenon was investigated by Zhang *et al.* [151] for 12, 15 and 20 wt% concentrations. Their results show that composites with nanofibrous interlayers electrospun from 15 and 20 wt% solutions show similar toughening effects – 92 and 87% increment in initial fracture toughness, 34 and 37% increment in fracture toughness for steady-state crack propagation compared to the multiaxial carbon fibre/epoxy reference composite – which exceed the effect of the 12 wt% concentration case – 55 and 16% increment. However, the 20 wt% concentration case demonstrated a more significant decrease in flexural modulus than the lower concentrations.

In terms of morphology, PCL nanofibres undergo polymerization-induced phase separation with epoxy, forming ductile thermoplastic-rich microphases at the delamination plane, which is crucial to interlayer toughening [151]. The positive effects of PCL interleaving can be further enhanced by decorating the PCL nanofibres with carbon nanotubes (CNTs) [154]. Research was conducted related to the combined application of PCL and polyamide 6.6 (PA6.6) as well, thereby enhancing both Mode I and Mode II fracture toughness – by 21 and 56% – of UD glass/epoxy prepreg-based composites. Meanwhile, the application of only PA6.6 nanofibrous interlayers enhanced the Mode I fracture toughness by 68%, but its effect on Mode II fracture toughness was negligible [153]. Marino *et al.* [155] achieved pseudo-ductile behaviour with the introduction of electrospun polyamide 6 (PA6) nanofibrous interleaves to interlayer hybrid composites consisting of prepreg-based UD carbon/epoxy and UD S-glass/epoxy layers. Nanofibrous interleaves were able to increase the fracture toughness of the composite and modify the occurring damage modes by suppressing delamination and inducing fragmentation. The experienced material behaviour was dependent on the production parameters of the different nanofibrous interlayer materials, such as the PA6 concentration of the solution – formed with a formic-acid and acetic-acid solvent mixture – used during the electrospinning process and areal density (Figure 6).

4.2. Thermoplastic film/pattern interlayers

Thermoplastic polymer interlayers can be applied in the form of films or structures with a designed pattern, which is usually produced by additive manufacturing methods. The use of thermoplastic interlayers has shown great potential in improving fracture toughness and damage tolerance of fibre-reinforced composites and ensuring pseudo-ductile material behaviour. Furthermore, these interleaves can make the composites mendable. High temperature and pressure

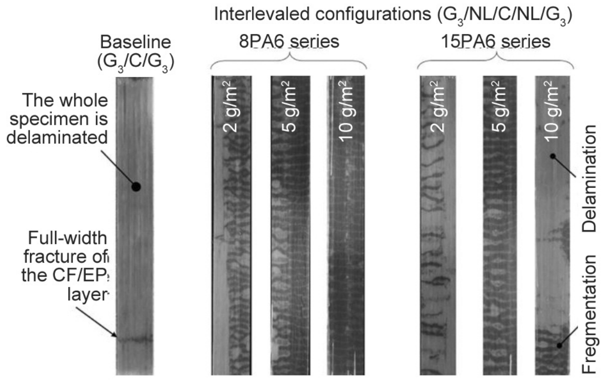

Figure 6. Hybrid composite specimens showing different types of damage (G – glass fibre/epoxy (GF/EP) layer, C – carbon fibre/epoxy (CF/EP) layer, NL – nanofibrous layer, 8PA6 or 15PA6– nanofibrous interlayer was electrospun from a solution with 8 or 15 wt% PA6 concentration) [155] (figure licensed under the Creative Commons Attribution 4.0 International License (CC BY 4.0)).

can enable the thermoplastic polymer to melt and flow into cracks, thus filling the gaps in the matrix cracks and delaminated areas [15, 156–158].

Marino and Czél [157] applied PA12 films as interlayer material in discontinuous carbon/epoxy – continuous glass/epoxy hybrid composites with a pseudo-ductile behaviour based on stable delamination initiated from designed discontinuities. Their results show that PA12 film interleaving increased the Mode II interlaminar fracture toughness by 100%, thereby increasing delamination resistance. Furthermore, the introduction of PA12 interleaves proved to be simple and effective in repairing failed composite specimens. The method requires the determination of the suitable repair parameters. The results showed that the mechanical properties of the investigated specimens can be restored by a repair cycle carried out in an autoclave at 185 °C temperature and at least 0.7 MPa pressure.

Zhang and Yasaee [158] investigated the impact of thermoplastic interleaves on the mechanical performance of open-hole notched quasi-isotropic ($[45/90/-45/0]_s$) CFRP composites, which usually fail by delamination at the 0°/–45° interface. They experienced that the applied interleaves were able to slow down delamination propagation, preventing early fibre fracture in the 0° plies. Besides, thermoplastic interleaves proved to be effective in repairing drill-induced damage.

It is also possible to introduce discontinuous interleaves as crack arrest features – *e.g.*, thermoplastic films – to suppress delamination propagation by increased fracture toughness in the modified regions. The technique proved to be effective in a research series carried out by Yasaee and coworkers [159, 160]. This method might contribute to a more controlled and designed failure process and may be suitable for restricting delamination damage into a predefined region.

A crack arrest feature was demonstrated as well by Anthony and coworkers [161, 162] in carbon/epoxy prepreg-based composites with an overlapped finger-jointed architecture using polyethersulfone (PES) and PES/EP hybrid interleaves. Their results showed increased strain-to-failure and delayed catastrophic failure of the composite realised by the interleaving.

Additive manufacturing has great potential for designing the interlayer of composite materials by allowing freedom in interlayer geometry. The application of PCL can be a combined method of adhesion modification and interlayer manipulation. It can create so-called weakened adhesion zones with a modified interface between the matrix and the reinforcement fibre. These zones can influence crack propagation by creating localised damage, mainly local debonding and delamination, and delay the fibre breakage, compared to the case of good interfacial adhesion, where cracks reaching the interface can cause the failure of fibres, as shown in Figure 7. These phenomena can result in pseudo-ductile behaviour with a gradual failure and higher energy absorption capacity [163].

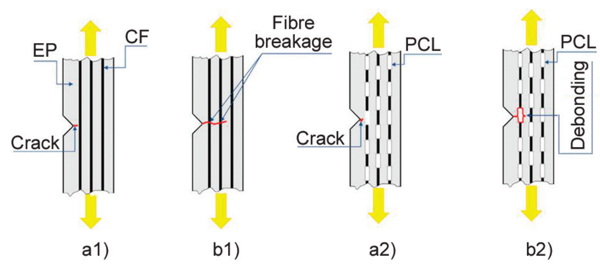

Figure 7. Crack initiation and propagation in reference and modified composites. a1) Crack propagation in reference composite with good interfacial adhesion; b1) propagating crack reaches the fibre and leads to its fracture; a2) crack propagation in composite modified with PCL interlayer; b2) in the interfacially engineered case, the cracks do not spread to the fibres, causing local damage [163] (figure licensed under the Creative Commons Attribution 4.0 International License (CC BY-NC-ND 4.0)).

The use of PCL as interlayer material was the focus of further research. Beylergil [164] applied 3D-printed PCL grids as interlayer material on the surface of carbon fabrics in order to increase the toughness of carbon/epoxy composites. Mode I opening tests showed that the thermoplastic layer caused gradual damage in the force-displacement curves, unlike the reference samples, which experienced sudden force drops. The modified samples continued to absorb energy after reaching peak load, leading to a significant increase in Mode I fracture toughness.

Magyar *et al.* [17] investigated the effect of PCL interlayer patterns on the failure behaviour of UD carbon/epoxy composites with ENF tests, three-point bending tests and Charpy impact tests. The results of the study indicate that PCL interlayer patterns lead to altered crack propagation with more energy dissipation and increased reliability by a gradual failure process. The failure behaviour and the determined properties are greatly influenced by the interlayer content. In the case of Charpy impact tests, the interlayer pattern was able to influence the damage mode and position as well; this enables the fundamental design of the failure process. In addition, a PCL interlayer pattern proved to be an effective method for providing the composites with repairability.

5. Conclusions

There is a significant need to influence and control the unfavourable, often catastrophic and unpredictable failure of composites, thereby increasing their reliability. Therefore, several research works have aimed to modify the damage processes and the failure behaviour of composite materials through the application of a wide range of methods. These methods can be based on different mechanisms.

Damage and failure behaviour can be controlled with a predefined failure location, which can be realized by introducing artificial discontinuities. However, most techniques aim to increase the toughness of the composite or even modify the occurring damage modes and achieve a pseudo-ductile behaviour demonstrated by a non-linear stress-strain response and a gradual failure of the composite. The toughening of the composite material can be realized through the modification of the matrix material, the reinforcement material, the fibre-matrix interface, or the interlayer. Furthermore, even artificial damage with proper design may be able to induce pseudo-ductility. In addition, a predefined failure mode can be combined with a predefined failure location, resulting in a designed failure.

Each method contributes to the desired failure behaviour by improving several properties, which determine the damage and failure process of the composite or by predetermining the location of failure. However, the different techniques have their own compromises, mainly in terms of some mechanical properties such as strength and modulus. While choosing the suitable method for controlling the failure behaviour of composites, the advantages and disadvantages of the available methods have to be taken into consideration.

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