

TRENDS ON REINFORCED POLYMER COMPOSITES – A REVIEW

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ABSTRACT

In recent years, the interest in using fiber reinforced composites (FRC) has increased due to their potential to replace traditional materials in various applications. The advantages of polymer composites from natural fibres are: biodegradability, high performance, market availability and low price. This article is based on a review that discusses both polymer composite materials and typical applications by other authors on fiber reinforced composites (FRCs).

KEYWORDS: polymer, fibres, composites

1. Introduction

The development of composite materials plays a crucial role in the advancement of technology [1-5]. Considering that both industrial and technological requirements are continuously increasing in fields (energy, naval, aerospace and automotive), there is a need for devices based on high-performance materials that can perform more than one function. This frequently involves the ability to respond in a controllable way to a certain physical or chemical stimulus, changing at least one of the material properties in direct correlation with the external stimulus [6].

Fiber reinforced composite materials are used in various industries due to their mechanical strength, modulus and corrosion resistance. Polymer nanocomposites based on carbon nanostructures for strain and temperature self-sensing have been proposed as the next generation of materials that are expected to overcome the well-known problems of inorganic sensors, such as complex processing, brittleness, narrow sensing range, and high sensitivity [7-10]. The polymer blend is the addition of two polymers at the macromolecular level, and the phase-separated morphology will influence the mechanical performance of the blend. Due to the poor reactivity of polymer-based materials with modifiers, poor adhesion between the epoxy and the modifier can result, and that can lead to a reduction in the mechanical properties of the composite. Therefore, the characterization of polymer matrix composites is essential [11].

For the synthesis of polymer-based composites there are, at least, two processes that are governing

the formation of polymers and associated composites: direct processing and in situ preparation. The direct mixing method involves synthesizing the desired nanomaterial and then dispersing it into the polymer matrix either by solution or mechanical dispersion. In the case of preparing composites based on in situ polymerization, the polymeric material creates a microenvironment to produce the desired metal/metal oxide from the precursors through series of chemical reactions. This process is of particular interest because the particle size and morphology could be easily controlled [12]. Analysing current trends, it can be seen that fiber reinforced thermosetting composites are among the most important materials. Due to the reliable mechanical performance of fiber reinforced polymer (FRP) composites, they have gained a huge market opening with inherent properties such as high corrosion resistance, high strength and improved fatigue resistance. These properties are superior because they are based on the interfacial bonds and cross-links between the matrix and the fiber. Thermosetting matrices such as epoxy, vinyl ester and polyester are commonly used as polymer matrices. Among them, epoxy resin is commercialized as a structural adhesive and as a polymer matrix for FRP composites [13]. Modification of epoxy resin (EP) with nanofillers has been extensively explored over the past three decades. Nanomaterials including carbon nanotubes, nanofibers (VGCF), organoclay, graphene oxide (GO) and its derivatives, functionalized aramid nanofibers (fANF) have been used in EP modification [14].

2. Kevlar fibres

Due to its unique properties, Kevlar fiber has become very popular as reinforcing material in composite materials, and its applications are considerably increasing [15-17]. Composite materials reinforced with synthetic fibres have become significantly popular over the years [18-20]. Among the synthetic fibres, poly-aramid fibres known as Kevlar fibres are mainly used for applications in industrial and advanced technologies such as ballistic armour, helicopter blades, pneumatic fittings, sports goods [21-23]. Their application has become considerably very wide due to its excellent mechanical properties, lighter weight, unique flexibility, corrosion resistance, ease of manufacture, etc. [24-26]. Compared with other synthetic fibres, Kevlar fiber has an elongation of fibres significantly lower and higher tensile strength and modulus [27]. They also exhibit very good properties at high temperatures for a polymeric material [28-30]. The glass transition temperature of Kevlar fiber is around 360 °C and it does not melt like nylon [31-33].

3. Polymer matrix composites (PMCs)

PMCs are promising materials for many engineering applications due to their superior properties such as self-lubricating properties, excellent chemical stability, electrical insulation properties, and wear resistance [34-37]. They have applicability in the aerospace, marine and sports industries. Also, other applications include the toy industry, coal handling equipment in power plants, bearings, transmission pumps, cosmetics and medical devices [38-42]. The advantages of polymer composites from natural fibres are: biodegradability, high performance, availability on the market and low price [43-48]. Hybrid composite is the material made from natural fibres and carbon fibres [49-51]. Interest in fabric-reinforced thermoplastic matrix composites has increased in recent years due to their faster processing possibilities compared to traditional thermo-resistant matrix composites [52, 53]. Textiles pre-impregnated with thermoplastic resins can be heated above the melting point of the matrix in seconds using infrared heaters and can be formed into a three-dimensional (3D) component [54-62], which involves significantly longer cycle times shorter than autoclave processing or resin transfer techniques [63-72]. Advanced fabrication techniques such as local reinforcement by tape placement is also facilitated due to the presence of the thermoplastic matrix [34]. Improved performance, shorter cycle times and reduced density are some features that have made woven thermoplastic composites attractive for the

aerospace and automotive industries [73-76]. Polymer matrix composites can be fabricated by various methods such as liquid casting, compression moulding, resin injection, and injection moulding, all of which belong to autoclave manufacturing methods [77-79]. In the field of external strengthening and repair of existing structures made of traditional materials such as concrete and masonry, fiber reinforced polymer (FRP) [80-84], has attracted much attention worldwide due to its ease of application, ability to limit impact aesthetic of repair works to the original structure and its appropriate reversibility for historical or artistic reasons [85-89]. Currently, almost all FRP applications [90-92], involve FRP made with glass fiber (GFRP) [93-96], carbon fiber (CFRP) [97-102], and in a smaller measure aramid fiber (AFRP) [103-117].

4. Epoxy resin

There are different types of epoxy resins, such as bisphenol A diglycidyl ether, cycloaliphatic epoxy resins, trifunctional epoxy resins, and tetrafunctional epoxy resins [118]. The global commercialization of epoxy resins market reached USD 25.8 billion in 2018, and for the year 2022 it is estimated to be around USD 34 billion [119]. Among these, bisphenol A diglycidyl ether is the most widely used in the coating industry due to its low cost and many desirable properties such as hardness, gloss and chemical resistance. Epoxy resin is one of the most common thermoset resins widely used as composite matrix and high-performance coatings due to its excellent characteristics such as: mechanical strength, chemical resistance, low shrinkage and electrical insulating properties [120-122]. Since the 1950s, various reports have appeared in the literature detailing the complex internal nanostructure of epoxy resin [123]. One of the disadvantages of epoxy resin is the brittleness caused by the tight three-dimensional (3D) polymer network. In recent years, efforts have been made to solve the problem of fragility. The hardness of epoxy can be improved by using elastomers, thermoplastic materials, nanofillers [124]. The most common component of epoxy resins is diglycidyl ether bisphenol A (DGEBA). DGEBA is industrially synthesized by reacting bisphenol A (BPA) with a large excess of epichlorohydrin under alkaline conditions [125]. The behavior of the resin depends on certain factors such as time, temperature, curing speed and pressure on the curing process. Characterization of the cure-dependent thermo-mechanical behavior of a fast-curing resin under process-relevant conditions remains a challenge [126-128]. It is known that when properly cured, epoxy resins exhibit a high crosslink density. When used as matrices in polymer composites, brittle resins can

produce composites with low damage tolerance [129-131]. Epoxy resins are frequently used as matrix of fiber reinforced composites. Matrix cracking has been shown to occur throughout the fracture process of the composite, particularly under fatigue loading. Brittleness of epoxy resins is the main source of composite failure, which can trigger composite defects such as delamination and debonding [132].

5. General presentation of the applications of composite materials

Cheng Chen *et al.* [133] performed a comparative analysis of natural fiber reinforced polymer and carbon fiber reinforced polymer for the strengthening of reinforced concrete beams. Due to the large amount of impregnated epoxy resin, the cost advantage of natural fibres was offset, and the overall cost efficiency of NFRP laminates ranged from 60% to 160% compared to CFRP laminates.

Sebastian Huayameres *et al.* [134] used unidirectional glass fiber reinforced epoxy, carbon fiber reinforced quasi-isotropic epoxy, and quasi-isotropic glass fiber reinforced epoxy to compare three-point bending and torsion methods to determine the viscoelastic properties of fiber-reinforced epoxy. This study showed that an irregular sample width can result in large dispersions in storage modulus values. Torsion tests, on the other hand, gave results that were consistent for fiber-reinforced composites regardless of specimen length, providing a more suitable method if materials are scarce and specimen length must be limited. Absolute values for viscoelastic properties cannot be directly compared between the three-point bending and torsion methods. At three-point bending, the storage and loss moduli were always higher than those measured in the torsional modulus for all of the studied composites.

This demonstrates that three-point bending measures higher moduli than torsion, regardless of fiber orientation, fiber type, or epoxy resin used for the two devices and test modes specified in this study.

Abdel-Hamid I. Mourad *et al.* [135] investigated the effect of nano additives on the damage resistance of a newly developed Kevlar fabric. Three types of nano additives were investigated: (1) silicon carbide (SiC), (2) aluminium oxide (Al₂O₃), and (3) carbon nanotubes. Damage was mainly observed in and around the impact area. The addition of small amounts of nano additives to Kevlar composites effectively improved the damage propagation resistance and interlaminar shear strength of the fabricated composites. In addition, increasing the nano additive content an improvement in the energy absorption capacity of the composites, especially with SiC and Al₂O₃ is observed, as the number of damaged

layers and the percentage of damaged area decreases. Among all the samples examined, the lowest number of damaged layers with the smallest damaged area was obtained with the addition of 0.5% by mass.

Ali Tabatabaeian, and Ahmad Reza Santhosh G. and Rajath N. Rao [136] fabricated polymer hybrid composites based on thermosetting resins using different hybrid fibres/fabrics with and without castor oil. The effect of reinforcements and/or castor oil on mechanical and thermal behaviours were evaluated. The results showed that the tensile, flexural and impact strength of composites with castor oil increases compared to composites without castor oil. The thermal behaviour was evaluated using DMA, the results showed an increase in the damping properties of the composites with castor oil. Lower glass transition temperature for castor oil composites indicated better damping behavior at higher temperatures.

Bo Yang *et al.* [137] analysed the nesting effect on laminates. They found that compared to increasing the length of the main flow channel, decreasing the width is a more significant factor in reducing through-thickness permeability. The results also show that a considerable degree of Nesting can occur due to unidirectional displacement, the total thickness can decrease by up to 5-6%, and the through-thickness permeability reduction can reach up to 80%. Bidirectional fabric displacement leads to a greater degree of Nesting, the total thickness decreases by more than 12%, and the through-thickness permeability decreases by more than two orders of magnitude.

Dhanush Kumar *et al.* [138] analysed the tensile behavior of Kevlar and glass fiber reinforced hybrid polymer composite as a function of cut-out size and position. Based on the experimental results obtained after subjecting the Kevlar and fiberglass reinforced composites to tensile testing, the authors state that the strength of the plates was improved by making the holes further from the centre, where the load carrying capacity was higher and the concentration factor upon request it was smaller. The authors stated that by moving the holes away from the centre a higher load capacity is obtained. It was also observed that the strain hardening coefficient increases linearly from sample 1 to sample 5 indicating higher resistance to necking before fracture. This means that the boards can be stretched longer by moving the holes away from the centre.

Silvio Leonardo Valença *et al.* [139] evaluated the mechanical behavior of epoxy composite reinforced with plain Kevlar fabric and glass/Kevlar hybrid fabric. Thanks to the values obtained after the mechanical tests, it can be seen that the structures developed with Kevlar and glass fiber hybrid fabrics have transferred the highest values of mechanical

strength and specific stiffness, becoming a new alternative for use as a structural laminated composite in the industrial market.

Xu W., *et al.* [140] performed experimental and analytical characterizations of finite interlaminar crack growth of 2D woven textile composites. The authors concluded that: the interlaminar static fracture resistance of the woven composite material determined by the double compliance method is consistent with that of the ASTM methods. The relative difference between the average fracture resistance obtained by these methods is less than 3%. The crack growth length can be accurately calculated using the dual compliance method, this advantage will significantly simplify the measurement of the strength at break of woven textile composites. The fracture strength of the unstable crack is given, in this paper, by using the dual compliance method. The fracture strength during unstable crack growth is lower than the initial fracture strength and results in the unstable growth of finite cracks.

Velmurugan V., *et al.* [141] performed an experimental evaluation of the mechanical properties of natural fiber reinforced polymer composites. The authors found that jute reinforcement with nylon fillers exhibits higher flexural strength compared to the combination of nylon and spider silk fillers in the flexural test. Reinforcement of the composite with spider silk and nylon fillers with epoxy matrix achieved higher toughness, and reinforcement with jute and nylon fillers with epoxy matrix achieved higher flexural and tensile strengths.

Murugan R., *et al.* [142] investigated the static and dynamic mechanical properties of glass epoxy composite fabrics and hybrid carbon composite laminates. Carbon laminate has higher mechanical strengths than glass laminate, except impact resistance. The variation in tensile strength and impact strength between the hybrid laminates is minimal, and the H2 hybrid layout has a higher flexural strength than the H1 hybrid laminate. The glass transition temperature, T_g of the H2 laminate has been changed by 5 °C to the glass laminate, which facilitates the higher operating temperature. The hybrid laminate with carbon fiber as the wrap layer, H2, performs better than another hybrid arrangement H1 and proves to be a good alternative for the glass laminate.

Jun Misumia and Toshiyuki Oyama [143] fabricated low-viscosity, high-strength epoxy resin modified by *in situ* radical polymerization method to improve the mechanical properties of carbon fiber reinforced plastics. In order to obtain both good mechanical properties, including the strength and toughness of the cured resin and the reduced viscosity of the uncured resin composition, the *radical in situ polymerization method* was applied to the epoxy resin

for the CFRP matrix. The results demonstrated that the *in situ radical polymerization method* can be effective for improving the fracture strength of CFRP laminates while maintaining the low viscosity of the uncured resin composition.

Mawarnie Ismail, *et al.* [24] studied the mechanical properties of composites made by reinforcing epoxy resin with bidirectional glass fibres and short RH fibres. The total fiber content for each sample was 30% and 70% by mass for the epoxy resin. The mass distributions for the RH fiber were between 5%, 10% and 15% by mass, while 25%, 20% and 15% by mass for the bidirectional glass fiber. It was indicated that its mechanical properties tend to decrease when the RH fiber content exceeds 5% by mass.

Kiran M. D., *et al.* [144] evaluated the breaking strength of carbon fiber and epoxy composite with different carbon fiber thicknesses. The composites were fabricated using hand-stretching technique by infusing 200 gsm, 400 gsm resin and hybrid carbon fabric laminate. The fracture toughness of the hybrid carbon fiber composites was studied using the single-edge notched beam method at room temperature (25 °C). From the experimental results, it was found that the epoxy composites reinforced with 200 gsm carbon fiber resist at breaking better compared to the composites reinforced with other fibres.

Goli E., *et al.* [145] developed a homogenized thermochemical model to simulate the production of unidirectional composites made of carbon fibres embedded in a thermoset dicyclopentadiene (DCPD) matrix using frontal polymerization (FP). The reaction-diffusion model is then solved using the finite element method to investigate the temperature evolution and degree of hardening during the manufacturing process. The results reveal two different processing regimes: At lower fiber volume fractions, the polymerization front speed increases with fiber volume fraction due to the increase in the effective thermal conductivity of the composite. At higher fiber volume fractions, the frontal velocity decreases with increasing fiber content due to the reduced heat source generated by the exothermic reaction.

Dai S., *et al.* [146] fabricated six types of 3D woven composites from carbon fiber and epoxy matrix. They studied the influence of fiber architecture on the tensile, compressive and flexural behavior of 3D woven composites. Four orthogonal weaves and two blocking angles were tested with the primary loading direction parallel to the warp direction. The mechanical performance was found to be affected by the distribution of resin-rich regions and the waviness of the load-bearing fibres, which were determined by the fiber architectures. Bonding points in resin-rich regions were found to be the sites

of damage initiation in all fabric types under all loading conditions, which was confirmed with both visual observation and image correlation strain maps.

In [147], the authors investigated a new epoxy resin matrix carbon fiber reinforced composite with viscoelastic sandwich layers by co-reinforcement technology. After testing, it was found that the addition of 0.1 mm thick damping film in laminates could lead to a 1.34% decrease in tensile strength and a 1.30% increase in flexural strength of the composites, reaching up to at 878.5 MPa and 766.3 MPa. However, the bending strength decreased with increasing thickness of the damping layer. The new co-reinforced damping composite exhibited high shear strength. The co-reinforced damping composites studied in this paper exhibited excellent damping properties.

In [148], the authors conducted a study that provides a low-cost solution to improve the mechanical and electrical properties of carbon fiber reinforced polymer composites by incorporating ultra-thin sheets of carbon nanotubes (CNTs) between CFRP layers (laminates). For this purpose, dry carbon fiber fabrics are first sandwiched between CNT sheets. The fabrics are then stacked and infused with epoxy to form a CFRP with interlaminated CNT sheets. Unlike the typical approach where micron-sized long CNTs are randomly distributed in a CFRP for reinforcement, this study uses 100nm-thick CNT sheets made of aligned and ultra-long (0.3 mm) nanotubes. Despite their negligible mass fraction of only 0.016%, interlaminar CNT sheets enhanced CFRP flexural strength by 49%, interlaminar shear strength by 30%, and mode I fracture strength by 30%. X-ray micro-tomography has shown that samples with interlaminar CNTs are significantly resistant to delamination and crack propagation. Moreover, the in-plane electrical conductivity of these composites increased proportionally with the mass fraction of CNTs, giving a maximum improvement of 278% over the reference sample for 0.048 mass% of CNT sheets.

6. Conclusions

- composite materials show a wide interest from researchers who approach a problem that starts from the matrix, reaches the reinforcement and the characterization of the materials;

- a lot of research considers composites with polymer matrix - either thermoset or thermoplastic;

- working models and interpretations of the results are proposed and, within certain limits, they are acceptable;

- many studies are carried out that take into account small variations of the matrix properties to identify the response of the composite;

- many studies refer to the matrix-reinforcement interphase (or other elements immersed in the polymer) to increase the utility value of the composites;

- we did not find bibliographic references regarding the analysis of the properties of polymer junctions, although in the design of the properties of composites a solution is to use layered matrices;

- some studies carried out at CCDCOMT aimed at the analysis of composites with a layered matrix consisting of the same polymer but modified in a different way for certain depths in the composite (the modifications being produced either by the introduction of solvents or by the addition of organic or inorganic agents).

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