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A Critical Review on Nanoparticle Filled Adhesives for Structural Applications

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Abstract

The objective of this review paper is to highlight some of the noteworthy research that has been done on the use of nanoparticles (NPs) to improve the performance of adhesively bonded joints (ABJs) against delamination initiation and propagation. Various nanoparticle applications, such as carbon-based, ceramic-based, and mineral-based nanoparticles, are covered. Interlaminar shear strength, fracture toughness, and fracture energy are the major parameters that have been considered for enhancing FRP delamination and fatigue resistance. The reported results indicatethat the inclusion of NPs in polymeric matrices leads to improvement of various material properties, even though some discrepancies in the results have been noted. Notwithstanding, additional research is required to address some of the issues that have not yet been tackled.

Keywords: *Nanoparticles, adhesives, structural application.*

1.0 Introduction

Adhesively bonded joints (ABJs) are made up of at least two similar or dissimilar adherends separated by a layer of adhesive, adhesively bonded joints have been widely used in various areas of technology, including the automotive, marine, space, and aircraft sectors, the adhesive is considered to be very important, as it is the agent that facilitates the transfer of load from one adherend to another adherend [1].

The aforementioned characteristics are the major factors that have created significant interest in ABJs across numerous industries. However, there are several limitations to utilizing adhesives as a joining agent, such as a long curing time, low-temperature resistance, and difficulties in disassembly. As a result, various studies [2] have focused on altering the characteristics of adhesives in order to address such shortcomings. The use of nanostructured reinforcements as a promising technique for improving adhesive response and overcoming some of the above-mentioned inadequacies is one of the recent initiatives made by various researchers.

Recently, new technology has emerged that has considerable potential for improving the mechanical performance of structural adhesives. Specifically, by forming a nanophase structure in the polymeric adhesive, consisting of small stiff particles or fibers with a diameter of about 5 to 50 nm [3].

A nano-particle (NP) is defined as a particle with a diameter of less than 100 nanometres. In fact, an NP can be considered as a link between atomic and bulk sizes. The following are the primary advantages of nano-size materials [4]: high specific surface area, high surface energy, fewer structural flaws, and physically separate physical properties from bulk materials. Unlike bulk materials, which have size-independent properties, NPs have size-dependent properties. NPs of various types have been created using a variety of materials. Each type of NP has one or more distinct physical qualities that are impossible to accomplish at the macro scale. Many researchers have been inspired by these unique

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qualities to focus their efforts on modifying the desired properties of adhesives using various types of NPs. The following elements should be considered: the adhesive's goal qualities, its compatibility with NPs, the NPs' functioning, and the service circumstances [5]. As a result, the first step should be to gather relevant information on the properties of each NP type and how they might alter the properties of the adhesive generated by their inclusion. Because the most popular NPs used in reinforcing adhesives are classified by type, therefore, a brief description of each group of NPs is discussed below.

2.0 Different Types of NPs

2.1. Carbon Nano Tubes (CNTs)

Carbonnanofibres (CNF) have been increasingly used as reinforcing materials in recent years. Composites with high performance, high modulus and strength, excellent stiffness, and creep resistance are all advantages of CF [5-6]. Despite these benefits, the mechanical characteristics of CF/thermoplastic composites are inadequate because CF has weak interfacial adhesion with most thermoplastic polymers due to its non-polar surface. Yu [7] demonstrated that a composite of MWCNTs and epoxy resin has good fracture toughness and fatigue strength. Jojibabu et al. [8] investigated the rheological characteristics, thermal stability, and lap shear strength of epoxy adhesive joints reinforced with MWCNTs, graphene nanoplatelets (GNP), and single-walled carbon nanotubes. Wernik and Meguid [9] demonstrated that CNTs in the range of 1.0–1.5 wt per cent improved the mechanical characteristics of CNT-reinforced epoxy adhesives. An electrical-impedance approach was proposed by Kim et al. [10] to identify the surface imperfection in an adhesive joint. They demonstrated that the electrical impedance approach employing carbon nanotubes is a promising tool for detecting various sorts of bonding defects. Prolongo et al. [11] investigated the modification of an epoxy adhesive by the addition of varying contents of CNFs for bonding carbon fibre = epoxy composite adherends. They discovered that adding nanoreinforcing blocks of cement had no effect on the lap shear strength of single-lap joints.The dispersion of nanoreinforcements in the adhesive minimizes stress concentrations and improves stress distribution uniformity. In comparison with bulk specimens, nano-reinforced adhesives showed an increase in fracture toughness [12-14].

According to the experimental results by Razavi [15], the addition of mixed nano-particles improved the mechanical properties including the shear strength and elongation at failure. The incorporation of mixed SNPs and MWCNTs into the adhesive layer was found to have no degrading effect on the mechanical properties in the three studied weight percentages of nano-particles. Functionalized and non-functionalized MWCNTs were utilized by Fereidoon et al. [16] to increase the shear strength of SLJs according to ASTM-D5868-01. The average shear strength improved by 40.5 per cent when 1.5 wt% MWCNTs were added to the epoxy adhesive.

To improve the adhesive joint strength, May et al. [17] used inorganic nano-particles such as MWCNTs and Al_2O_3as as reinforcements to the epoxy/sol-gel adhesive. They discovered that including $Al_2O_3$and MWCNTs at weight percentages of 0.71 per cent and 0.05 per cent, respectively, resulted in higher lap shear strength than single phase-reinforced and unreinforced adhesives. The increase in lap shear strength was attributed to improved adhesive-to-substrate adhesion as well as adhesive cross-linking (cohesive strength).

2.2. Nanosilica (SiO_2)

Nanosilica contains porous, broad surfaces with a lot of hydroxyl groups and unsaturated residual bonds. Nano silica has been shown to be an efficient reinforcement for polymers in terms of increasing strength, flexibility, and durability. Furthermore, nano-silicahas been employed as an additive in high-performance and self-compacting concrete to improve workability and strength [18]. Kinloch et al. [19] convincingly established that adding low concentrations of nano-silica particles to a conventional rubber-toughened adhesive based on a two-part epoxy formulation boosts the adhesive's toughness significantly, as well as the glass transition temperature and single-overlap shear strength.

Zhou et al. [20] reported that single lap-shear experiments under quasi-static and cyclic loadings were used to investigate the impact of nano-silica on the adhesive characteristics of epoxy. Under quasi-static loadings, the adhesive strength of the compound was found to be 20% higher than that of neat epoxy. The benefit of having nano-silica in neat epoxy on adhesive joint strength was sustained even after hygrothermal treatment.

Hsieh et al [21] reported that the addition of silica nano-particles resulted in a considerable rise in the nanocomposites' fracture energy. Incorporating 6.0 wt% silica nano-particles resulted in a 36 per cent increase in fracture energy when compared to the neat epoxy polymer.

Metallic NPs have piqued the interest of scientists for more than a century, owing to their vast potential uses in nanotechnology and the recent boom in their use in biological sciences and engineering [22]. These materials can now be produced and modified to provide a variety of chemical functions. Metallic nanomaterials can be formed from most metallic elements of the periodic table such as nano-gold, nano-silver, and metal oxides (e.g., alumina (Al_2O_3) or zirconia (ZrO_2)) NPs

2.3 Alumina (Al_2O_3)

Alumina particles are a typical filler used to improve the thermal conductivity of adhesives, especially insulation adhesives. For adhesives intended to be used as an electrical or thermal channel, aluminium and silver powders or flakes are utilised to improve thermal and electrical conductivities. To achieve adequate conductivity, the filler volume content level is critical. Excessive filler content, on the other hand, may cause the adhesives' mechanical properties to deteriorate. Wetzel et al [23] has also shown that the addition of 13 nm diameter alumina particles increased the fracture toughness of epoxy cured using a cycloaliphatic amine, from 0.5 to 1.2 MPa.

The adhesive was modified with 5 or 10 wt% of either 50 nm diameter alumina nano-particles or alumina nano-fibers with a diameter of 2–4 nm and an aspect ratio ranging from tens to hundreds. Both of the nano-modifiers increased the peel and shear strength. However, the more successful modification was the addition of 5% of alumina nano-particles, which increased the peel strength of the adhesive by 50%, and the lap-shear strength by 15% [24]. Nano-sized alumina particles of various sizes and shapes were used in the development of epoxy-based film adhesives. These nano-scale components were mixed with an adhesive at a rate of 5 to 10% by weight. The results of the bonded aluminium samples showed that the additions increased the adhesives' peel and shear strength in almost all cases.

2.4 Nano Silver (Ag)

The incorporation of nano-particles into epoxy-resins offers environmentally benign solutions to enhancing the integrity and durability of coatings since the fine particles dispersed in coatings canfill cavities. The usefulness of nano-particles brings many advantages and opportunities to the paint and coating industry [25].

Necati Ataberk [26] reported that the strength of the adhesive and the strength of the joint is the different characteristics of an adhesive material. Maximum tensile strength (96.34 MPa) of the bulk specimen was obtained for 15 wt% Cu nano-particle added specimen, but the maximum joint strength (15.52 MPa) was obtained for 10% Cu nano-particle added specimen. After the SLJ tests, almost all of the failures occurred by adhesively (fracture between the adhesive and adherent). This can be commented as because the perfect integration between the adhesive and adherents cannot be reached, or the strength of the composite adhesive is higher than the linking between the adhesive and adherents' surfaces.

2.5 Carbon nano-fibers

An excellent adhesive is well known for having a high wettability across the adherend. The measurement of the contact angle is usually used to determine this behaviour. The lower the contact angle is better will be the wettability on the substrate. The effect of adding CNFs to an epoxy adhesive on its contact angle was studied by Prolongo et al. [27] used a carbon fibre epoxy laminate as the adherend. The measurement of contact angle is based on the adhesive type and substrate, but it is also reliant on the properties of the adherends' surfaces, such as their surface energy, roughness, and so on.

Several studies have been published on the effect of incorporating carbon nano-fibers into adhesives in order to improve joint strength and toughness. The tensile strength of joints bonded with a commercial epoxy adhesive enhanced with carbon nano-fibers was investigated by Xu et al.[28]. A variety of adherends were used, ranging from aluminum to poly(methyl methacrylate) (PMMA).The tensile strengths of Al/Al joints bonded with nano-fiber reinforced adhesives are all lower than those of neat epoxy joints. The strength of PMMA/PMMA joints bonded with nano-reinforced adhesives with varying fibre weight contents, ranging from 0.3 to 0.8 per cent, however, exceeds that of the joint bonded with non-modified adhesive the most significant rise was up to 30%.

The effect of adding carbon nano-fibers to epoxy adhesives on the lap shear strength of joints of carbon fiber/epoxy laminates was studied by Prolongo et al [29]. Apart from improving the mechanical properties of nano-reinforced adhesives, the filler's small size could improve adherence to the substrate, resulting in new anchor sites. This mechanism could be enhanced when theadherend is a composite of epoxy matrix reinforced with long fibers due to the high chemical compatibility with the epoxy adhesive reinforced with carbon nano-fibers.

Thao Gibson et al.[30] found similar results after studying the development of epoxy-based adhesives manufactured with coated and uncoated vapor-grown carbon nano-fibers. They found that when CNFs were added to the adhesive, the shear strength of metal-metal and composite-metal couplings remained constant. This modification, however, resulted in a significant decrease in electrical resistivity and an increase in thermal conductivity.

2.6 Graphite nano-particles (GNP)

GNP nano-particles have outperformed carbon nano-tubes in terms of fracture mitigation. Rafiee et al. [31] found that using as little as 0.1 wt per cent of GNPs, epoxy nano-composites had a 53 per cent increase in mode I and a 126 per cent rise in mode-I ILFS, compared to 20 per cent and 66 per cent, respectively, when MWCNTs were used. In another study, Rafiee et al. [32] also showed the superiority of GNP as an effective enhancer of mode I fracture toughness compared to nanoclay. A similar trend was observed by Chandrasekaran et al. [33], who demonstrated that the inclusion of thermally reduced graphene oxide (TRGO) led to better performances in comparison with GNPs.

Ahmadi et al. [34] also highlighted the benefits of functionalization of GNP particles on the fracture toughness enhancement for all modes I, II, and III of GF/EP laminates. The authors studied functionalization agents: NH2, graphene oxide, NH2, and G-Si yielded the best results, with significant improvement of all three modes of fracture toughness values.

Walker et al. [35] introduced 1.5 vol % GNP particles in bulk silicon nitride ceramics, which is even more brittle than thermoset polymers, and obtained a 235% improvement in the fracture toughness of the ceramic. The crack deviation was shown to be the mechanism accommodating the observed dramatic increase.

2.7 Other types of NPs

Other forms of nano-particles have also been studied for their impact on fracture toughness and strength. The mode I ILFT of CF/EP laminates reinforced by soft rubber NPs and rigid silica-NPs was investigated by Zeng et al. [36]. They observed the effect of different types of NPs as well as their mixed state. Rubber NPs were 2.5 times more effective than silica NPs in enhancing the ILFT.

Moreover, by combining both NP types, the margin of improvement fell in between the margins obtained by using the individual NP types. Backed by scanning electron microscopy (SEM) analysis of the delaminated surfaces, the authors attributed the improvement of interlaminar toughness to the cavitation of nano-rubber particles, void growth, and debonding of nano-silica from the epoxy matrix. However, they mentioned that the hybrid effect of nano-particles needed further investigation. Hsieh et al. [37] also mixed rubber and silica nano-particles and observed that the inclusion of rubber NPs led to higher fracture toughness than could be attained by silica NPs. Other similar studies regarding nano-silica particles can be found in references.

Kelkar et al. [38] used functionalized and un-functionalized alumina nano-particles as interleaf materials, and reported, respectively, 51% and 74% increase in the mode I fracture toughness compared to baseline specimens.

Zheng et al. [39] reinforced a less commonly used polymer, polysulfone, with CNTs and used the mixture for interleaving CFRP laminates. They reported an increase in values of both mode I and mode II fracture toughness's, with the optimum CNT concentration of 10 wt%. Yan et al. [40] incorporated graphene into natural rubber and observed that at low fatigue strains, the crack growth was accelerated with the addition of NPs, while the opposite effect was reported for higher strains. The authors suggested this behaviour to be linked to the crystallization process. At higher strain rates, crystallization occurs, causing a deviation of the crack path and leading to higher energy dissipation, therefore reducing crack growth. The inclusion of graphene seemed to exaggerate this phenomenon

3.0 Conclusions

The focus of this review paper has been to highlight the role of NPs in enhancing the joint strength of bonded joints, in turn improving the inter laminar shear strength (ILSS) of the laminate composites and adhesively bonded joints created by such reinforced resins. As seen, a significant amount of work has been conducted on the noted topics, leading to a large number of publications. The conclusion of these works can be summed up by stating that the inclusion of a small number of appropriate nano-particles in a polymer matrix can lead to significant improvements in the mechanical properties of the hosting matrix, so long as the NPs have been dispersed uniformly within the matrix. A few works have also reported the detrimental influence of this strategy, by observing degradation in delamination resistance of FRPs, especially under impact loading, when the matrix ductility was actually reduced due to NP content above the appropriate threshold limit.

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