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Single lap joints bonded with structural adhesives reinforced with a mixture of silica nanoparticles and multi walled carbon nanotubes

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ABSTRACT

The average shear strength and elongation at failure of adhesively bonded single lap joints (SLJs) were studied when different mixtures of two nanoparticles i.e. silica nanoparticles (SNPs) and multi walled carbon nanotubes (MWCNTs) were added to the adhesive. The experimental results showed that adding the mixed nanoparticles had a significant effect on the mechanical behavior of SLJs. The highest improvements in the shear strength and elongation at failure among different weight percentages of the investigated mixed nanoparticles were 28% and 36%, respectively, which were related to 0.8 wt% of the mixed nanoparticles. Moreover, the improvement of mechanical properties due to addition of the mixed MWCNTs and SNPs was higher than the improvements obtained for corresponding single-type nanoparticles with the same weight percentages. The fracture surfaces and damage mechanisms of single lap adhesive joints were also investigated and a correlation between the joint strengths and the fracture surfaces was found.

1. Introduction

Adhesively bonded joints (ABJs) provide many advantages compared to traditional joints such as welding, bolting and riveting. Excellent resistance to mechanical vibration and fatigue loads, weight and cost savings, sealing ability and minimum local stress concentration are some advantages of adhesive bonding over other joining techniques. However, these joints suffer from some disadvantages such as long curing time, poor temperature and humidity resistance and requiring careful surface preparation before bonding. In order to improve the mechanical behavior of adhesive joints and composites, researchers have proposed several methods such as adding nano and micro particles and macro/micro fibers into the adhesive layer and composite matrix. For instance, Fereidoon et al. [1] used functionalized and non-functionalized MWCNTs to improve the shear strength of SLJs according to ASTM-D5868-01. The results revealed that the average shear strength increased up to 40.5% by adding 1.5 wt% of MWCNTs into the epoxy adhesive. Also, they showed that the bonding surface of aluminum substrates was not chemically affected by MWCNTs. May et al. [2] added inorganic nanoparticles including MWCNTs and $\gamma - Al_2O_3$ as reinforcements into the epoxy/sol-gel adhesive to improve the adhesive joint strength. They showed that incorporation of $\gamma - Al_2O_3$ and MWCNTs with weight percentages of 0.71% and 0.05%, respectively, resulted higher lap shear strength compared to the corresponding single phase-reinforced and unreinforced adhesives. The increase of lap shear strength was attributed to the improvement of adhesion between the adhesive and substrates and adhesive cross-linking (cohesive strength). Tutunchi et al. [3] used silica nanoparticles into an acrylic adhesive to enhance the shear strength of steel-glass/epoxy composite joints. They showed that addition of silica nanoparticles up to the weight percentage of 1.5 led to increase the shear strength by about 29%. A significant decrease in water contact angle and consequently increase in interfacial wettability of the acrylic adhesive with steel and glass/epoxy substrate was also reported and therefore, the shear strength of adhesive joints was increased. Zhai et al. [4,5] investigated the effect of adding nano alumina particles into an epoxy adhesive on improving the adhesive strength using pull-off adhesion test. By increasing the nano alumina content, the adhesive strength gradually increased and reached to a maximum value at a weight percentage of 2. Improvement of adhesive strength caused the failure locus to change from interfacial to combination of interfacial and cohesive. Cui et al. [6] used second phase nanoparticles namely nano-hexagonal boron nitride (BN) into a mixture of adhesive resin and micro silver particles to improve the shear strength and electrical conductivity. The results showed that addition of 3 wt% of BN considerably increased the electrical conductivity and shear strength of the adhesive. Dorigato et al. [7] synthesized zirconia

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nanoparticles into an epoxy adhesive by sol-gel technique in order to improve the shear strength of the adhesive joints. The results revealed that the introduction of zirconia nanoparticles into the epoxy adhesive led to significant improvement of shear strength by about 60% at optimal filler content of 1 vol%. The presence of zirconia nanoparticles caused the interfacial wettability and chemical compatibility between the adhesive and substrate to increase as evidenced from equilibrium water contact angle. Pinto et al. [8] incorporated graphene nanoplates (GNPs) into the polyvinyl acetate as an adhesive to improve the shear strength of the adhesive joints. The results showed that 0.1 wt% of GNPs content increased the shear strength more than 50%. They also found that addition of GNPs into the adhesive caused an increase in the equilibrium angle of contact due to nanofiller hydrophobicity. This meant that increase of shear strength of the adhesive joints could be attributed to strong interfacial interactions between the polymer chains and GNPs. Chen et al. [9] investigated the effect of silica nanoparticles on the adhesive mechanical properties as a function of silica nanoparticle contents. They observed that the adhesive strength and fracture toughness increased by about 30% with the nanoparticle weight percentage of less than 10 and then decreased by further increasing the silica nanoparticle content. Tang et al. [10] used MWCNTs and spherical particles such as rigid nanosilica and soft submicron-rubber particles to produce hybrid system and single system nanocomposites. They studied the effect of incorporation of these nanoparticles on mechanical properties with special concentration on the fracture toughness, electrical conductivity and glass transition temperature of the epoxy/nanocomposites. The results showed that the hybrid-reinforcement system improved all properties compared with the single-reinforcement system. Kinloch et al. [11] studied the effect of nanosilica particles on the mechanical and thermal properties of a typical rubber adhesive. They demonstrated that adding nanoparticles with low weight percentages led to significant increase in the adhesive toughness, shear strength and glass transition temperature. Chen et al. [12] used two different silica nanoparticle sizes of 12 nm and 100 nm in the EPON epoxy matrix to study the effect of silica nanoparticle sizes and weight fractions on the tensile strength. The results showed that the tensile strength of composites was independent of the size and weight fraction of silica nanoparticles. Wang et al. [13] investigated the effect of adding nanosilica particles into the starch-based wood adhesive on improvement in the shear strength and water resistance. The highest increases of shear strength of the adhesive joints in dry and wet states were obtained as 50.1% and 84%, respectively, while, a maximum water resistance improvement of 20.2% was obtained for the reinforced adhesive. Gojny et al. [14] studied the effect of as-received and amino functionalized double-walled carbon nanotubes (DWCNTs) on improving the mechanical properties of epoxy nanocomposite by a standard calendering technique. The results showed that improvements in the studied mechanical properties including the tensile strength, the Young's modulus, the strain to failure and the fracture toughness occurred only in the matrix reinforced by amino functionalized DWCNTs at weight percentage of 0.1. Khan et al. [15] investigated the effect of graphen nanoplates on the mechanical properties of polyvinyl acetate (PVAc) adhesives. They observed that adding 0.1 vol% graphen nanoplates into the neat adhesive caused an increase of 100% in the tensile strength and 50% in the stiffness of the reinforced adhesive joints. Moreover, the maximum increases in the adhesive toughness and strength were found as 7 and 4 times, respectively, compared to the neat adhesive. An et al. [16] employed carbon nanotubes functionalized with ozone treatment for oxidation followed by chemical reaction polyethyleneimine for depositing onto the carbon-fiber fabric using electrophoretic deposition technique. Significant shear strength and fracture toughness improvement of carbon-epoxy composites using CNT treatment were reported. Hsieh et al. [17] incorporated MWCNTs into an epoxy polymer to investigate the modulus, the fracture energy and the fatigue performance of the modified polymer. Highest improvement in the mentioned mechanical properties was reported at weight percentage of 0.5. Wood et al. [18] showed that overlap glass-glass joints bonded with an adhesive reinforced by silica nanoparticles improved the adhesion bond between the adhesive and glass substrate. The highest shear strength of the adhesive joint failed cohesively occurred at 0.5 wt% as a result of improving the adhesion bond between the adhesive and adherends.

Similar researches on the effect of second phase addition to the adhesive layer were carried out by other researchers [19–28].

The present study is focused on the shear strength and elongation at failure of SLJs reinforced with two types of nanoparticles i.e. silica nanoparticles and multi-walled carbon nanotubes under quasi-static loading. The experimental results of nanoparticle reinforcement are compared to the corresponding single-phase reinforcements and unreinforced adhesive joints and the effect of types and weight percentages of the nanoparticles are investigated on the mechanical behavior of the adhesive joints.

2. Experimental

2.1. Adhesive material and nanoparticle dispersion procedure

In order to reinforce the epoxy based adhesive, a mixture of two nanoparticles including the spherical silica nanoparticles (TECNANNANOMAT, Los Arcos-Navarra, Spain) [29] and multi-walled carbon nanotubes (NanoAmor, Los Alamos, New Mexico, USA) [30] with equal weight ratio (1:1) were added to the adhesive layer. The material properties of the nanoparticles are presented in Tables 1 and 2. The surface treatment of SNPs was not carried out in this study. The adhesive joints were manufactured using a bi-component epoxy adhesive named UHU plus endfest 300 (UHU GmbH & Co. KG, Bühl, Germany) with mixing ratio of 1:1 for hardener and binder (see Table 3). In order to separate and fully disperse the nanoparticles inside the adhesive, the mechanical stirring followed by ultrasonic sonication using Bandelin-SONOPULS HD-3200 were used. This machine is capable to generate high-performance ultrasound with high intensities and ultrasonic amplitudes which are transferred into the liquid through working tools known as probe. Selecting a proper sonication time for the dispersion process of the mixture of nanoparticles was of significant importance, such that under or over level energy can lead to deviation from optimum mechanical properties of adhesive joints so that beside visual checking of the nanoparticles dispersion into the resin, researchers have recommended several methods such as using SEM images (see Figs. 1 and 2) and measuring the mixture viscosity which were used in this study. The viscosity of the epoxy adhesive reinforced by studied nanoparticles was measured to evaluate the dispersion state of nanoparticles in the adhesive. The optimum dispersion of nanoparticles is obtained when the viscosity of suspension reaches to a peak. We used this method and obtained the optimum sonication time for the adhesive joints reinforced by MWCNTs and silica nanoparticles. Three different weight percentages in the mixture of nanoparticles containing MWCNTs and SNPs with equal weight ratio were added into the adhesive. The weight percentages of 0.1, 0.25 and 0.4 were considered for each constitutive nanoparticle giving rise to 0.2, 0.5 and 0.8 wt% of combined nanoparticles in total. First, the mechanical stirring was carried out at 100 rpm for 15 min and in the meantime MWCNTs were added into the adhesive. Subsequently, for three weight percentages of 0.1,

Table 1
Properties of SNPs [29].

Silica nanoparticles	
Average particle size	10–15 (nm)
Specific surface area (SSA)	180–270 (m^2/g)
Density	2.2 (g/cc)
Purity	99.999%