

Minireview Article

Research progress on mechanical properties of modified epoxy resin reinforced by fillers

ABSTRACT

In this paper, the research progress of different types of fillers (carbon nanomaterials, ceramics, metals, natural fillers) to enhance the mechanical properties of epoxy resin in recent years was reviewed, and the enhancement effect of filler types and dispersion on the mechanical properties of epoxy resin was summarized, and the development direction of epoxy resin enhancement modification was prospected.

Keywords: Epoxy resin, packing type, packing dispersion, mechanical properties

1. INTRODUCTION

Epoxy resin (EP) is a thermosetting polymer used in the preparation of composite materials and is known for its excellent properties, such as excellent mechanical and thermal properties, as well as resistance to corrosive solutions and corrosive environments [1-4]. In addition, the epoxy resin can also be modified by adding appropriate reinforcing materials to obtain the desired properties [5]. As a kind of crosslinked polymer, epoxy resin has high brittleness due to its high crosslinking after curing [6], Low fracture toughness [7], Peeling strength difference [8] and other disadvantages, Its application under high load and strong impact conditions is limited [9]. Therefore, enhancing the mechanical properties of EP to meet the required application requirements has been the focus of research.

At this stage, it is necessary to enhance the mechanical properties of EP without sacrificing other excellent properties, and it is generally modified by adding specific fillers, using the volume effect and surface effect of the filler to achieve the combination with epoxy resin, so as to achieve the effect of enhancing the mechanical properties of EP [10, 11]. In addition, the study found that the factors affecting the properties of EP nanocomposites include the original properties of the filler and the dispersion of the filler in the EP matrix, so modifying the surface of the filler to improve its interface with the matrix has become a major research direction. This paper will focus on the research status and progress of different types of fillers to enhance the mechanical properties of EP.

2 ENHANCEMENT OF DIFFERENT TYPES OF FILLERS

The addition of filler is one of the most common methods to enhance the mechanical properties of EP matrix, and its high specific surface area will increase the interface interaction between it and the matrix. Even at low fill content, its introduction into the matrix phase significantly enhances the performance of EP, so the use of fillers in EP is a way to enhance the other properties of EP without reducing the excellent properties of EP itself. At present, researchers have successfully enhanced the mechanical properties of EP by using carbon nanofillers, ceramics, metals and natural fillers.

2.1 Carbon nanofiller

Compared with traditional metal structures, carbon filler-based polymer nanocomposites are popular because of their superior specific strength, higher thermal stability, and better fatigue and corrosion resistance [12, 13]. Therefore, carbon nanofillers are a better way to strengthen epoxy resin composites, and many studies are devoted to adding carbon microfillers and nanofillers to epoxy resin composites: graphene oxide, carbon nanotubes, carbon nanofibers, etc [14].

Alipour et al. [15] Various concentrations of graphene nanosheets (GNP), ranging from 0 to 0.7 wt%, were studied using an effective solvent approach to strengthen flax/epoxy composites, thereby correlating the formation of GNP networks with the degree of reinforcement of the resulting nanocomposites. The microstructure characteristics characterized by wide X-ray diffraction (WAX), scanning electron microscopy (SEM), and transmission electron microscopy (TEM) determined the correlation between GNP enhancement efficiency, nanolayer network formation, and layer number. TEM analysis illustrates that as GNP content changes, the nanolayer distribution changes from a regional distribution to an effective bridging network, helping to maximize tensile and bending properties at 0.5wt % GNP. This was supported by morphological observations developed by SEM, confirming various enhancement mechanisms due to GNP networks leading to enhanced interfacial adhesion between fibers and substrates. Similarly, with the formation of the network, the crack length at the fiber-matrix interface and the crack density in the nanocomposite (as determined by optical microscope images of the tensile test sample) decreased significantly. TEM observations also demonstrate that the plane-to-plane and edge-to-edge contact areas of GNP have a crucial influence on the reinforcement efficiency of nanocomposites. Above the optimal content (0.5 wt%), the load transfer capacity caused by GNPs network formation is adversely affected by GNPs planar contact, ultimately leading to a significant reduction in mechanical properties.

The addition of a separate graphene filler will make the matrix properties have certain limitations, such as easy aggregation, so filling with other related materials of the graphene family is currently a common method to enhance EP properties. Qu et al [16] The epoxy resin was modified with 0.0-0.5wt % graphene oxide (GO). The mechanical properties at room temperature (RT) and low temperature (77 K) of carbon fiber /GO modified epoxy resin composites prepared by hot pressing process were systematically studied. The results show that the good dispersion of GO in epoxy resin leads to the enhancement of interfacial adhesion between carbon fiber and epoxy resin matrix. It was observed that the low temperature mechanical properties of CF/ epoxy resin composites were significantly enhanced by the introduction of GO. It is found that the optimal GO content is 0.2wt %, which can effectively improve the low temperature bending and interlayer shear strength of carbon fiber reinforced epoxy resin composites. In addition, the results show that the mechanical properties of CFRP composites at 77 K are higher than those at room temperature. Therefore, the modification of epoxy resin by introducing GO is a promising means to achieve high and low temperature mechanical properties of CFRP composites.

Chanda et al. [17] The effects of the arrangement, structure, concentration and loading conditions of carbon nanofibers on the nonlinear stress-strain behavior of epoxy nanocomposites were studied. The disordered CNF/epoxy nanocomposites were prepared and tested at different strain rates. The elastic modulus and tensile strength of randomly arranged and transversely arranged nanocomposites increase with the increase of CNF concentration. Compared with the randomly arranged nanocomposites, the transversely arranged nanocomposites have lower modulus and strength.

Carbon nanotubes (CNTs) are considered as excellent reinforcement materials for the preparation of high-performance composites because of their unique bonding structure, high tensile strength and high elongation at break [18]. Most current studies on CNTs/EP composites have focused on improving the mechanical properties of EP by adding different types of CNTs fillers and making them well dispersed in EP resins. Seyaji et al [19] Carbon nanotube (CNT) fibers were treated with acid and modified by epoxy resin impregnation. The impurities in CNT fibers were reduced by acid treatment, and carboxyl functional groups were formed on the surface of CNT, which had a positive effect on the strength of CNT. However, some defects can occur on the nanotube surface, which negatively affects the elastic modulus. During epoxy penetration, the pores of the carbon nanotube fibers are filled with epoxy, thereby improving the interfacial interactions between the carbon nanotube bundles, or interlocking through polymer chains that wind around the tubes, or even by forming covalent bonds between the carboxy-functionalized nanotubes. By comparing the mechanical properties of the epoxy/carbon nanotube fibers before and after acid treatment, it was found that the final modulus of the fibers was reduced when the fibers were treated with acid before epoxy resin infiltration. The modulus of the epoxy/carbon nanotube fiber reaches 103 N/tex, which is about 20% higher than that of the acid-treated epoxy/carbon nanotube fiber. The acid treatment prior to the penetration of the epoxy resin produced functional groups on the CNTs surface that enhanced the strength of the fibers through a strong reaction between the epoxy chain and the CNTs. On the other hand, structural defects imposed by acid on CNTs resulted in a decrease in their elastic modulus.

Babahan-Bircan, et al. [20] A novel bio-based epoxy amine coating and its nanocomposite coating derivatives graphene, carbon nanotubes (CNTs) and fullerenes were prepared as substitutes for BPA for the first time, and their thermal and mechanical properties were compared. Epoxy functionalized tung oil (ETO) was prepared by Diels Alder reaction. As a curing process, primary amine hardener (Jeffamine D2000) is used at four different temperatures from 25 ° C to 150 ° C. Epoxy amine nanocomposites were prepared by adding carbon nanoparticles (graphene, carbon nanotubes and fullerenes) with 1:1 molar ratio of epoxy: amine. The cured coatings were analyzed by IR, DSC, TGA and gel content tests. According to TGA analysis, the new coatings have a thermal stability of up to 430°C, and these cured films show two glass transition points (T_gs) by DSC. However, of the three nanocomposites, the graphene system had the largest T_g in both phases, while the carbon nanotube system had higher T_s and the lowest weight loss at 800 ° C, with a residual of up to 60%. The mechanical properties of the cured coating were measured by pendulum hardness, pencil hardness, pull-off adhesion,

intersection adhesion, impact resistance and reverse impact resistance tests. All cured coatings have good mechanical properties in terms of hardness, cross adhesion and impact resistance. In general, the epoxy nano composite coating showed better thermal and mechanical properties than the epoxy nano composite coating.

2.2 Ceramic packing

Ceramic particles are widely used in polymer composites and erosion wear protection due to their excellent properties such as low shrinkage rate and high mechanical strength. Its high specific surface area and high activity enable it to form a force higher than the van der Waals force with the EP matrix at the interface. At the same time, due to its small elongation deformation under tensile stress, it can effectively hinder crack propagation, thus achieving the role of strengthening EP [21]. At present, enhancing the mechanical properties of EP by adding SiO₂, SiC, BN, Al₂O₃ and other ceramic fillers has been widely studied.

Li et al [22] In order to improve the bonding strength of water-based epoxy coating of carbon fiber reinforced polymer (CFRP) composite, micron silica particles modified by γ -aminopropyl triethoxysilane (γ -APS) were prepared by sol-gel method, and rough surface was established on CFRP surface. The surface morphology, roughness, chemical composition and wetting behavior of the coating were characterized by the pull-off adhesion test, and their effects on the adhesion of the coating were analyzed. The results show that the adhesion strength of the CFRP epoxy coating is increased by about 120% with the introduction of modified silica particles. Compared with the traditional sanding method of CFRP composites, this study provides a convenient and environmentally friendly method to achieve the adhesive strength requirements of water-based epoxy coatings without causing mechanical damage and dust pollution of the composites.

An et al [23] Two kinds of SiO₂@GO complexes (GO-A and GO-B) with different structures were obtained and characterized by electrostatic interaction and chemical graft modification of graphene oxide (GO) with silica (SiO₂). The results confirmed that both methods successfully grafted SiO₂ onto the surface of graphene oxide. The distribution of SiO₂ particles on the surface of GO-A is more dense and more agglomerative, while the distribution on the surface of GO-B is more uniform. Epoxy resin (EP)/ graphene oxide composites are then prepared. The curing mechanism of EP/GO composites was studied by differential scanning calorimetry and in situ infrared spectroscopy. The results of tensile test, hardness test, dynamic mechanical analysis and dielectric measurement show that EP/GO-B has the highest tensile property, with a tensile strength of 79 MPa, which is 43% higher than the original EP. In addition, the addition of fillers increased the hardness of EP, and the energy storage modulus of EP/GO-B was the highest, reaching 1900 MPa. The dielectric constant, volume resistivity and breakdown voltage of EP/GO composites were increased by the addition of hybrid fillers SiO₂@GO. EP/ GO-B has the lowest dielectric loss, better insulation performance, higher volume resistivity and breakdown voltage.

Kolipakula et al [24] The tribological properties of glass fiber reinforced plastic (GFRP) composites filled with Al₂O₃ and SiC powder particles were studied. Experiments were carried out on a pin-disk tribometer to evaluate the changes of friction and wear rate with normal load and sliding speed under different conditions. The glass-epoxy composites with and without fillers were subjected to sliding tests under normal load (20 N, 40 N, 60 N, 80 N, 100 N) and sliding speed (2.62 m/s, 4.18 m/s, 5.23 m/s, 6.85 m/s, 7.85 m/s), respectively. Metal GFRP composites were prepared by manual lamination method. The content of filler (SiC and Al₂O₃) in epoxy glass composites varies greatly. SiC is 0 ~ 10%, Al₂O₃ is 5%. The wear surfaces of glass-epoxy composites were studied by scanning electron microscopy. The wear of composite material (pin sample) was studied on a digital balance before and after the experiment. The results show that compared with epoxy fiber composites without metal powder, the prepared metal epoxy fiber composites have excellent wear resistance.

Luo et al [25] The thermal conductivity and mechanical properties of epoxy resin (EP) composites were improved by using polyether sulfone (PES) and silver nanoparticle functionalized boron nitride nanosheets (AgBNs). The 0D silver nanoparticles (AgNP) prepared by the reduction reaction of DMF and AgNO₃ have electrical conductivity, and the interface contact of two-dimensional boron nitride nanosheets (BNs) is increased, thus improving the thermal conductivity path of agbn. The results of SEM and optical microscopy showed that the three-dimensional continuous heat conduction path of agbn was constructed based on PES/EP bicontinuous phase structure, and it was improved with the increase of agbn content. The results show that the thermal conductivity of PES-EP /AgBNs reaches a maximum value of 0.54 W m⁻¹ K⁻¹ under a load of 10 wt% AgBNs. The thermal conductivity of EP/AgBNs (0.45 W m⁻¹ K⁻¹) and EP/BNs (0.36 W m⁻¹ K⁻¹) is much higher than that of EP/ AgBNS with the same filler content. At the same time, the fracture mode of PES-EP/ agbn gradually changed from brittle to ductile, and the tensile strength, flexural strength and fracture toughness of PES-EP/ agbn were superior to EP/ agbn. This method effectively reconciles the conflict between thermal conductivity and mechanical properties of epoxy resin composites, providing conceivable applications in high-performance thermal conductivity devices.

2.3 Metal packing

The metal and its oxide fillers have phase stability, thermal stability and high surface area, which can enhance the interface interaction between it and the epoxy resin matrix, thereby improving the mechanical properties of EP [26]. At present, in addition to silver, aluminum and its oxides, new two-dimensional materials have been developed, such as two-dimensional transition metal carbon (nitrogen) compounds (MXenes) and metal-organic frameworks (MOF).

MXenes are a new class of two-dimensional materials that have attracted much attention due to their excellent electronic properties, abundant surface ends and unique atomic structures. Wei et al [27] Epoxy composites were prepared by electrostatic self-assembly of electronegative $Ti_3C_2T_x$ nanosheets, electropositive polystyrene microspheres and sacrificial templates of different PS sizes. The thermal conductivity of the prepared FMX/EX composites varies with the size of the PS template. With a template diameter of 0.74 μm , the FMX-0.74/EX composite has the highest thermal conductivity (0.32 W/m·K), which is due to the greater contact between the $Ti_3C_2T_x$ nanosheets and the more complete microstructure of the $Ti_3C_2T_x$ skeleton. At the same time, under the protection of epoxy resin, the heat transfer performance of $Ti_3C_2T_x$ nanosheets remains stable at possible operating temperatures. FMX-0.74/EX also shows good thermal stability during heating and cooling cycles. This indicates that $Ti_3C_2T_x$ nanosheets have potential applications in the thermal management of electronic devices.

Haeri et al. [28] Environmentally friendly cerium-Organic framework-oriented graphene oxide (GO/Ce-TA MOF) was developed for the first time to improve the tensile stress and fracture toughness of epoxy groups. In addition, the mechanical and thermal stability of the CeTA MOF reinforced polymer coating was studied. The cross section of the epoxy composite shows the fracture morphology, which strongly proves the high toughness and uniform dispersion of ce based mof particles in the epoxy matrix. The heat resistance of Ce-TA MOF and GO/Ce-TA MOF reinforced coatings is approximately 20% and 30% higher, respectively, than that of pure epoxy. The Pull-off adhesion test results showed that the coating did not fall off after the introduction of Ce-TA MOF and GO/Ce-TA MOF into the epoxy coating. In addition, the impact test results show that adding additives to the epoxy matrix can satisfy the resistance of the coating to mechanical damage. The tensile test results show that the breakpoint energy of Ce-TA MOF and GO/Ce-TA MOF nanocomposites is increased by 275% and 195%, respectively, and the tensile strength exceeds 90% and 60%, respectively. The final results show that the mechanical properties of Ce-TA MOF can be considered to be more significant than GO/Ce-TA MOF particles.

2.3 Natural filler

Compared with traditional fillers, natural fillers have the advantages of easy availability, light weight, waste utilization and biodegradability. The results show that the thermal and mechanical properties of EP can be enhanced by adding various natural fillers into EP, which has been widely used in electronics and other fields.

Shah et al. [29] The mechanical properties and thermal conductivity of epoxy resin were enhanced by acacia particle modification. The effects of different AC particle wt.% load on curing and properties of epoxy resin were studied successfully. FTIR spectroscopic analysis of epoxy-reinforced AC particles showed the presence of chemical and hydrogen bond crosslinking in the composites. DSC results showed that AC particles accelerated the curing reaction of epoxy resin due to the tannin part of AC particles. When 1.0 wt.% AC particles were added to the epoxy resin, the toughness and impact strength were increased by 52% and 94%, respectively. Under the high load of AC particles, the toughness and impact strength values are lower than 1.0 wt.% of AC particles, but still higher than the corresponding values of pure epoxy resin. The thermal stability of AC-filled epoxy composite was also improved, and the thermal stability parameters showed a gradually increasing trend. Only when 2 wt.% AC particles were strengthened, the peak degradation temperature moved from 403°C/min to 442°C/min. The Tg values of 2 wt. % reinforced AC particle composites were observed by DSC and DMA at 143°C and 148°C, respectively. These values are slightly lower than the neat matrix values, and this decline may be due to the flexible chain of AC particles. SEM analysis shows that the phase distribution of AC particles is good, reflecting the shear yield and plastic deformation in a large volume. The prepared composite material is stable at high temperature in an inert atmosphere.

Nano-clay minerals have high hardness and are suitable for enhancing the structural properties of modified polymer matrix. Vo et al [30] An efficient synthesis method was reported to fill biological epoxy resin nanocomposites with highly exfoliated epoxy grafted montmorillonite. The binary nanocomposites (MMT-PGMA) synthesized by in-situ photoinduced polymerization of glyceryl methacrylate were covalently bonded to the bio-epoxy resin matrix, and the ternary nanocomposites (MMT-PGMA/ bio-epoxy) were designed in the presence of green polyamines as curing agent. The structure and morphology of the materials were characterized by FTIR, TGA, XR, SEM, TEM, etc. The results showed that the D-spacing between clay layers increased from 1.23 nm to more than 2.2 nm after graft. The glass transition temperature (Tg) of the bio-epoxy resin was significantly increased by ultrasonic treatment of the homogeneous solvent-free dispersion of the hybrid clay nano-filler. This can be explained by the good dispersion and chemical surface reactivity of the nanofillers, ensuring a strong interfacial adhesion to the matrix.

3. DISPERSION OF FILLERS IN EP

Zheng et al.[31] The effects of the type and particle size of various dielectric fillers on the anti-settling and viscosity reduction of the prepared suspension were studied by studying the dispersion of fillers on the electrical and mechanical properties of the embedded materials. The 0.3 μm BaTiO₃ prepared by hydrothermal method has the best effect in the preparation of medium suspension. The prepared ECM has high electrical and mechanical properties. The embedded capacitors of different sizes are further processed with a change rate of less than 10%. In order to verify the applicability of ECM in electronic products, reliability tests are also carried out. This study shows the importance of good dispersion of dielectric fillers in polymer matrix, and provides an effective method for selecting dielectric fillers suitable for dielectric machining.

Zanghellini et al [32] The various nano-fillers are dispersed directly into the tetrahydromethylphthalic anhydride hardener by ultrasound without the use of additional solvent systems. We analyzed the size distribution of the aggregates, characterized the rheological properties of the dispersions, such as estimates of long-term stability, sedimentation, and viscoelastic behavior, to evaluate the mass of the dispersions in depth, and compared these results with samples produced by trm. In particular, 0.5 wt% carbon oxide nanotubes showed the best results of all the US samples, revealing that the size distribution of most of the aggregates was comparable to that of the samples produced by trm, with a good combination with the hardener, forming a permeable network inside the dispersion, which points to stability against settlement and therefore appropriate long-term stability. Higher fill volumes showed similarly good results, but also formed a more pronounced internal network as well as larger clumps. CNF can also be successfully dispersed by the US method, but it is generally found that the dispersion of CNF shows a low probability of forming a long-term stable system.

Li et al. [33] In this paper, spherical particle combinations with different size scales are used to increase the load fraction of fillers in epoxy matrix composites. In this study, the multi-scale particle size of spherical Al₂O₃ particles achieves a high load of up to 79 vol%. The highest thermal conductivity of liquid epoxy resin filled with Al₂O₃ measured by steady-state method is 6.7 W m⁻¹ K⁻¹, than pure epoxy resin (0.28 W m⁻¹ K⁻¹). Three models based on Maxwell Mean Field Format (MMF), Differential effective Medium (DEM) and seepage theory model (PTM) were used to evaluate the thermal conductivity data we measured. We found that both DEM and PTM models give good results at high volume fractions. We also observed a significant (10-15%) reduction in thermal conductivity of the Al₂O₃ filled cured epoxy resin samples. We attribute this decrease to the increase in thermal interface resistance between Al₂O₃ particles and cured epoxy resin matrix caused by curing shrinkage during the reaction. Our experiments show that systems with multi-scale particle sizes have lower viscosity and can be filled with a higher proportion of fillers. Therefore, we expect that higher thermal conductivity (perhaps >12 W m⁻¹ K⁻¹) can be achieved in the future by filling spherical fillers with higher thermal conductivity (such as AlN, SiC), increasing the loading fraction by multi-scale dispersed fillers, and reducing the effect of curing shrinkage.

4. CONCLUSION AND OUTLOOK

With the continuous expansion of EP market demand, it is required to have excellent mechanical properties, adding filler to EP to enhance the modification has become the most direct method. In recent years, various types of fillers have been applied, researchers through modification, compounding and other methods in the premise of not destroying the excellent performance of EP, to complete the optimization of its specific performance, broaden its function and application range. Among them, carbon nano fillers, ceramics, metals and their oxides and natural fillers are widely used to enhance the mechanical properties of modified EP because of their excellent properties. In addition, some biopolymers based on EP have more application potential than commercial polymers due to the advantages of green environmental protection, low cost and improved mechanical properties. At present, there is still room for improvement in the enhancement of the mechanical properties of EP by fillers. In the future, the following aspects can be considered: (1) The addition amount of filler is a decisive factor in determining the mechanical properties of composite materials, and it is necessary to strengthen the research on the addition ratio of resin/filler and the uniform dispersion of filler in EP; (2) Deeply understand the mechanism of various fillers, and study the modification method of new fillers; (3) In-depth study of various types of mixed fillers synergistically enhance the mechanical properties of EP composites.

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