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Epoxy–Silica Functionally Graded Materials: A Review

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Abstract:– This article provides an overview of the studies that have been conducted on the characteristics of epoxy resins containing various types of silica nanoparticles and microparticles, as well as their performance in the industrial application of functionally graded materials (FGMs). Silica nanoparticles and microparticles are used to create epoxy resins in order to improve various properties, such as thermal stability, adhesiveness, electrical conductivity, strength, modulus, and toughness. This review examines the literature that has been published in the last decade, compares the results, focuses on the mechanical and thermal properties, and discusses the changes that have resulted in improvements in those properties. Previous experimental findings are presented and contrasted to demonstrate the extent to which silica filler content contributes to improving the properties of composite materials. The findings reveal that the characteristics of epoxy compounds can be improved by adding a particular amount of silica particles. There is a correlation between an increase in the silica amount and an increase in the Young modulus of epoxy compounds, this correlation becomes stronger as the silica amount increases. Additionally, the tensile strength of epoxy compounds increases to a certain limit as the amount of silica nanoparticles increases. In contrast, the hardness of the material increases as the silica amount increases. The density of the material also increases steadily as the silica amount in the material increases. According to thermal analysis results from calorimetric research on epoxy–silica systems, the glass transition temperature increases as the silica amount increases.

Keywords:– FGM, Epoxy, Silica nano/microparticles, Composites.

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1. Introduction

Functionally graded materials (FGMs) are composite materials that have a continuous variation in their structures through the thickness [1]. This results in a gradual and constant change in material characteristics from surface to surface, which can improve many different properties [2]. FGMs are used to eliminate sharp interfaces in composite materials, which can cause failure [3]. Functionally graded materials are a class of composite materials that have been created using a unique technique to accomplish qualities that monolithic materials cannot. Figure 1 (a), and (b) show that composites' qualities are evenly dispersed throughout the material, resulting in a homogeneous product [3, 4].

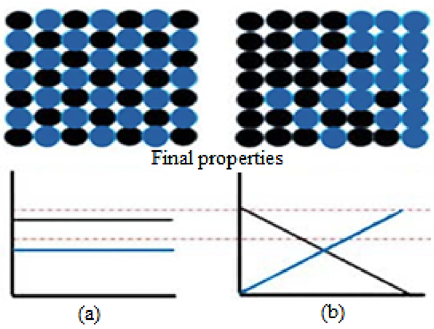

Fig. 1: Variation (a) composite and (b) FGM characteristics [1, 3].

Structural gradients were first proposed for polymers and composites in 1972 [3] to mimic natural materials including teeth, bones [5], and Bamboo [6]. The notion of functionally graded materials was developed in Japan by Niino in 1984 [4, 7–9]. The primary goal was to produce thermal barrier materials with increased mechanical capabilities and thermal resistance by gradually modifying compositions to endure significant temperature differences in turbine engines, fusion reactors, and space structures [4, 10].

Recently, FGMs widely used in medical equipment, biotechnology, electronics, power generation, nuclear reactors, transportation, military operations, and aerospace. These applications need thermo-mechanical and chemical stability [11]. Compositionally, FGMs can be divided into two categories: those that are continuous in nature, and those that are step-wise. Step-wise FGMs are multi-layered structures having an interface between the discrete layers, whereas continuous FGMs continuously change the properties and specifications of the material components. [8, 12] as illustrated in Fig. 2 (a) and (b).

An overview of techniques and manufacturing processes of functionally graded materials was published some years ago. Saiyathibrahim et al. [13] reviewed the process techniques of functionally graded materials between 2000 and 2012. Shailendra et al. [3] reviewed the manufacturing process of functionally graded materials between 1999 and 2012.

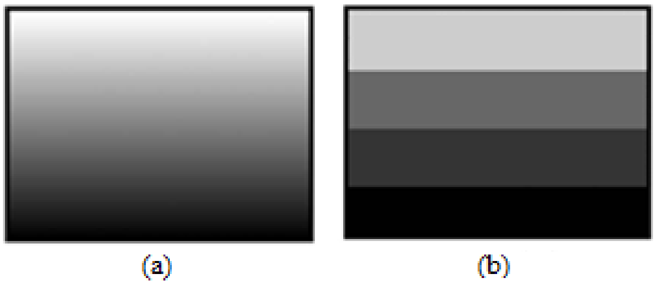

Fig. 2: Variation (a) composite and (b) FGM characteristics [1,3].

In addition, many researchers conducted experiments to examine the effects and consequences and toward meeting the challenges of FGM [9, 14, 15]. Numerous research studies have been conducted to produce ceramic-metal [7], alloy-ceramic [16], ceramic-ceramic [17], and alloy-alloy compositions, which are used to obtain graded materials with varying thermal resistance through the use of specialized fabrication procedures.

In this review, the literature produced in the last decade is analyzed, and the mechanisms underlying the improvement of the properties of polymer-based composites through the gradation of silica epoxy content in polymer-ceramic combinations are discussed. The addition of silica, carbon fiber, etc. not only improves the flexural strength and hardness of graded composites but also imparts thermal stability, which is highly desirable in industrial applications [18]. The paper will focus light on silica epoxy composition with a focus on mechanical and thermal properties and provide formulating guidelines. Everyone interested in the science and engineering of gradient materials for the composition of silica/epoxy composition is anticipated to benefit from this contribution.

2. Epoxy/silica characteristics and their industrial applications

2.1. Epoxy

Among their many uses, epoxy resins are found in everything from automotive and aerospace parts to ships and windmill blades to construction adhesives and coatings, epoxy resins are a common form of thermoset polymer [19–21]. The modification of epoxy resins and curing agents is documented, which describes the flame resistance, adhesive property, thermal property, and chemical stability of epoxy resins [15, 22–24]. Epoxy resin is filled with silica particles to increase the composites' thermal stability, curing behavior, thermal mechanical properties, and dynamic mechanical properties [25].

2.2. Silica

Silica occurs naturally. Inorganic and biogenic crystalline or nanocrystalline silica exist. Free silica forms meter-sized quartz crystals or amorphous masses of submicroscopic crystallites with diverse microstructures. No natural analogues for synthetic polymorphs exist [26]. In recent decades, silica has gained attention for its unique features and a vast range of technical applications. Silica is added to epoxy to improve thermal stability and mechanical characteristics. These low-cost components rarely affect or lower product price, but they can improve product quality [27–29]. Furthermore, the mechanical property improvements that can be accomplished with silicate-based fillers, like elasticity, flowability, and hardness have been the topic of several studies to increase their thermal, chemical, electrical resistance, dimensional stability, and mechanical properties [30–33].

3. Research and experiments in epoxy/silica fabrication and characterization

Epoxy resins need fillers. Silica fillers improve strength, modulus, toughness, and fatigue. Thus, the inclusion of a silica filler could enhance the performance of a variety of applications [34]. The work of Rihan and Abd El-Bary [35] focused on fabricating graded silica/epoxy by filling epoxy Novo Floor S12 of silica particles with median diameters of (45–250) μm to obtain a stepwise graded structure content of silica. The prepared materials were used as coating materials for the floors of chemical laboratories. The correlation between electrical properties and wear resistance with the variation of silica wt.% content in graded silica/epoxy composites was studied in different percentages (0, 10, 15, and 20 wt.%). The results showed that the wear resistance increased by increasing the wt.% content of silica. Electrical conductivity remains constant till about 15 wt.% content of silica but, it increases at a high rate for wt.% content of (15% – 20%) and above 20 wt.% content. It increases slowly with increasing silica content and breakdown voltage remains constant till about 15 wt.% content of silica, but it decreases at a high rate for wt.% content of (15% – 20%), and above 20 wt.% content, it decreases slowly with increasing silica content.

Majeed [20] prepared a composite material with an epoxy resin matrix and silica fume reinforcement. The addition of silica fume had a weight percentage of (0.5, 1.5, 2.5, 2.5, and 3 wt.%). Complying with ASTM, hardness and compressive tests were conducted. There was an increase in hardness with increasing levels of silica fume added up to 2%, after which the hardness decreased. Tests of compression strength revealed a rise in compressive with increasing silica fume ratios, up to 2 wt.%, after which the strength drops.

Tronoh silica sand grinding to nanoparticles using a dry ball mill was the subject of research by Ahmad and Mamat [36]. Nanoparticles of silica sand were utilized to develop and investigate the characterization of polymer-based composites. The tensile and flexural properties of the composites were measured, and the tensile and flexural strengths of polymer matrix composites containing up to 15% silica sand nanoparticles increased. The densities of polymer-based silica sand nanoparticle composites were also evaluated. With more silica sand nanoparticles, density increased. Silica is denser than polymer; therefore, adding it as reinforcement increases its density.

In a continuation of the work above, Ahmad et al. [37] presented research on the effects of nanoparticle silica dispersion in epoxy on composite mechanical, thermal, and physical properties. The addition of 15 wt.% silica nanoparticles to composites increases their elastic modulus, however increasing silica sand nanoparticles in epoxy reduced nanocomposites' crystallinity.

Ochi et al. [38] studied epoxy/silica hybrids. The epoxy resin contained bisphenol. The silane alkoxides were employed as inorganic sources. To make the epoxy/silica hybrid plates, the percent silica (0, 4, 6, 10, 14, and 19.8 wt.%) of the hybrid materials was assessed. Using an X-ray microanalyzer, the silica dispersion in the adhesive of the joints was studied. The hybrids containing silane alkoxides exhibited a significantly higher bonding strength for silicone rubber than the unmodified epoxy resin, however, the high bonding strength observed in the bisphenol/silane alkoxides hybrids was due to the formation of the interfacial bonding between the silica networks formed on the surface of the substrate and the epoxy networks in the adhesive layer.

Henk et al. [39] used the fumed nanoparticle silicon dioxide to filled epoxy both pure network polymers and polymer/particle composites have been tested for partial electrical discharge resistance. It has been observed increase in the performance of epoxy/SiO_2 regarding partial discharge resistance. Where nanoparticle silicon dioxide dispersed in epoxy increases endurance.

Zamanian et al. [40] studied the influence of interphase-particle adhesion and the interphase area surrounding nanoparticles on the stress distribution and mechanical characteristics of epoxy/silica nanocomposites. Analyzed and illustrated the effect of interfacial bonding conditions on Young's modulus of epoxy/silica nanocomposites, as well as the effect of interfacial adhesion on the tensile characteristics of nanocomposites. In comparison to the nanoparticle volume fraction, the results showed that complete bonding adhesion increased the Young's modulus of nanocomposites. In addition, by including nanoparticles, the rate of growth of the elastic moduli under poor bonding conditions was slowed. Where interfacial adhesion quality had a significant impact on nanocomposites' tensile performance. Clearly, the results demonstrated that increasing the interphase modulus stiffened the nanocomposites.

Yu and Kidane [41] discussed the effect of geometrical and unique elastic material parameters of every constituent and porosity on the effective elastic properties of FGM, and also the effect of individual material properties, to comprehend the effect of the specific elastic modulus on the effective elastic properties of FGM. A micromechanical model was used to predict the effective elastic characteristics of FGMs with much distinct heterogeneity. To compare the theory with the experiment results, the elastic characteristics of two-phase functionally graded silica spheres in a polymeric matrix were calculated. The effect of geometry and the elastic properties of each ingredient and void on the effective elastic properties of that material was investigated. It has been observed that the difference in elastic deformation between the homogeneity and the base material has a slight effect on the effective elastic properties. Consequently, the effective elastic modulus is strongly dependent on the weight percentage and material characteristics of each component. The numerical results for various FGMs were in excellent accord with the experimental results.

Shumiya et al. [42] investigated the application feasibility of FGM made from epoxy resins packed with crystal silica powder using an experimental and computational technique. Samples were filled with epoxy resins. Crystal silica has low permittivity and large filler particles, while TiO_2 rutile crystal has high permittivity and small filler particles. SiO_2 powder was $0.3 - 19.9$ μm (average: 5.89 μm) in diameter. TiO_2 powder has a $0.2 - 3.9$ μm (average: 0.67 μm) diameter. The investigation found the diameter of filler particles impacts the viscosity of base materials (lower diameter, higher viscosity). Filler particle precipitation depends on a balance between centrifugal force and base material viscosity.

Ibrahim et al. [43] examined the mechanical characteristics of silica-reinforced polymer composites. It was discovered that the increase in elastic modulus is proportional to the reinforcement material weight fraction.

Bakri et al. [44] explored the enhancement of epoxy nanocomposites with mesoporous silica/nano-silica. Tensile stress, elastic modulus, and yield stress were combined with mesoporous silica and nano-rigid silica's size, structure, and characteristics. Physical and mechanical qualities increased linearly with particle loading, unlike mesoporous particles or nanoparticles like rubber.

Battistella et al. [45] presented the findings of an experimental program designed to produce and characterize nanocomposites based on epoxy resin modified with fumed silica nanoparticles (0.1, 0.3, and 0.5 vol.%). The degree of resultant dispersion and interfacial adhesion were studied. Under mode loading, fracture toughness, and tensile properties were studied. The fracture toughness was significantly enhanced, and a trend toward the enhancement of the crack growth limit under fatigue was detected. The 3-Aminopropyl-modified silica particles nanocomposite exhibits the greatest improvement in failure strain. According to the result of a fracture toughness test, the primary effect of the nanofiller addition is an increase in fracture toughness with all nanocomposites manufactured.

AlSaadi and Erklig [46] studied the effect of silica nanoparticles on tensile, flexural, and mixed-mode fracture toughness. The inclusion of nanosilica altered the composite's strength, modulus, and fracture toughness, according to the findings. At 1.0 wt.% nanosilica, the highest increase in fracture toughness reaches 41.8 % and 35.7 % when compared to polyester.

Daramola and Akintayo [47] reinforced epoxy resin with 0.5 μm silica particles, 0.5, 1, 2, 3, 4, and 6 wt.% silica particles were added to an epoxy resin to make composites. The composite's mechanical properties (tensile, flexural, hardness) were tested. Silica particle incorporation improves elasticity, flexural modulus, and hardness with a 2% threshold. Adding silica decreased tensile and flexural strength, with the flexural strength values being slightly higher than their respective tensile values.

Ozcan et al. [48] incorporated silica nanoparticles and graphene nanoplatelets into an epoxy coating matrix to increase the mechanical properties of polymer composites. Adding graphene to epoxy boosted the coatings' flexibility and impact resistance by 8.3% and 157%, respectively, compared to neat epoxy. Silica/graphene nanohybrid increases microhardness by 53.8% and scratch resistance by 29.7%. Graphene and silica increased epoxy coatings' hardness and scratch resistance.

In another article, Yeasmin et al. [49] prepared the polymer composites-based epoxy with mesoporous silica, seven different types of silica Particle sizes (10 nm, 3 μm, 5 μm, 20 μm and 63 – 212 μm) were mixed for epoxy/silica nanocomposite and epoxy/silica micro-composites. Comparing the influence of mesoporous micro-silica pore size on filler-matrix interactions. Epoxy composites with tiny concentrations of mesoporous silica showed higher thermal performance than nano/micro-silica and epoxy composites. Utilizing differential scanning calorimetry, the thermal stability and phase transition temperature were evaluated. The silica with nanopores incorporated into epoxy resin demonstrated competitively superior interfacial performance results than modified nanosilica fillers. This was attributed to the fact that micro-silica fillers participated in gelation with the epoxy resin during the long-time curing steps, as a result of a higher crosslinking density than that of silica nanocomposites.

Kucharek et al. [50] examined the thermal conductivity and compressive characteristics of epoxy composites based on aerogel concentration and particle size. The results demonstrated that integrating aerogel particles into the resin might reduce density and thermal conductivity by more than 40%. However, silica/epoxy composites exhibited a linear trade-off between compressive characteristics and heat conductivity.

Lavorgna et al. [51] studied the effects of silica enrichment of carbon nanotubes on the thermal and thermal-mechanical properties of composite materials based on epoxy. The study revealed that silica nanoparticles collected around the walls and ends of Carbon nanotubes. The thermal, thermomechanical, and flammability properties of a nanocomposite comprising 1 wt.% of various fillers were determined and compared to those of epoxy resin in its pure state. The results demonstrated that the inclusion of filler raised both the rubbery and elastic modulus. Specifically, in the rubbery zone, the moduli of composites with silica-enriched carbon nanotubes were approximately 240 and 285% more than the modulus of a pure epoxy. In the glassy zone, a 25 % increase in the storage modulus was observed. With silica-enriched carbon nanotubes, the glass transition temperature rose by approximately 20 °C, and the damping behaviour was much improved.

4. Results and discussion

4.1. Density/volume change

Density and percent weight measurement of epoxy/silica (nanoparticles, fused silica powder, and silica powder with fabric composites). Figure 3 displays the findings of density tests conducted on epoxy mixtures containing nano silica, fused silica powder, and silica powder combined with fabric. Produced from data in [4, 37].

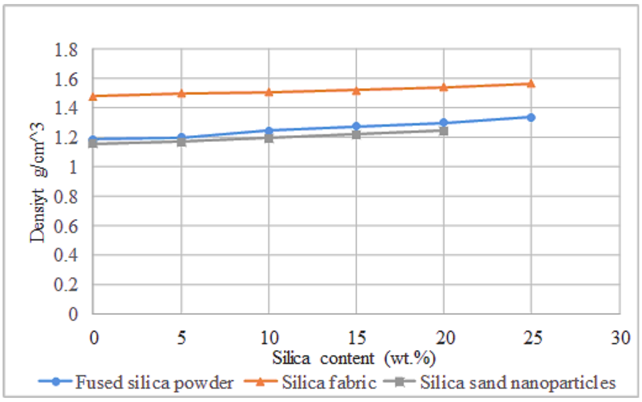

Fig. 3: Effect of weight percentage of different silica on the density of composite, recalculated from data in Reference [4, 37].

The graph in Fig. 3 demonstrates the relationship between density and weight percent silica for nano silica and silica samples with and without fabric. With increasing silica content, material density increases progressively. Composites including fabric have greater density than composites lacking fabric (only fused silica powder). Due to the bigger size of the silica particles, nano-composites of silica demonstrated a lesser increase in density when compared to composites containing fused silica [4, 37].

4.2. Mechanical behaviour of the epoxy-various types of silica composites

4.2.1. Hardness test

Hardness test results for epoxy with silica fume, silica fume with carbon fiber, and nanoparticles with carbon nanotube are displayed in Fig. 4, recalculated from data in [20, 52].

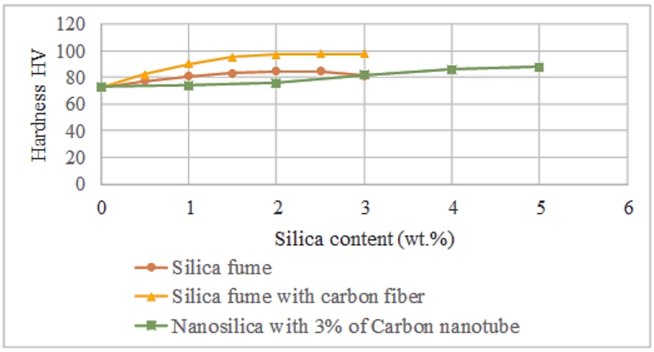

Fig. 4: Effect weight percentage of different silica on the hardness of composite, recalculated from data in [20, 52].

Hardness increases with increasing the ratios of silica fume and carbon fiber correspondingly, but with carbon fiber, it is greater than for silica fume and fixed at 2 wt.% and thereafter, where the carbon fiber compensated for the drop in mechanical properties. The hardness of silica nanocomposites increased as the ratio of nanoparticles to carbon nanotubes increased, although the increase was less than that of composites containing silica fume, which could be attributable to the extremely small size of nanoparticles [20, 52].

4.2.2. Young's modulus

Results of Young's modulus test for epoxy with nanosilica, fused silica powder, fine silica powder with fabric, and micro silica in Fig. 5. Produced from data in [4, 19, 43].

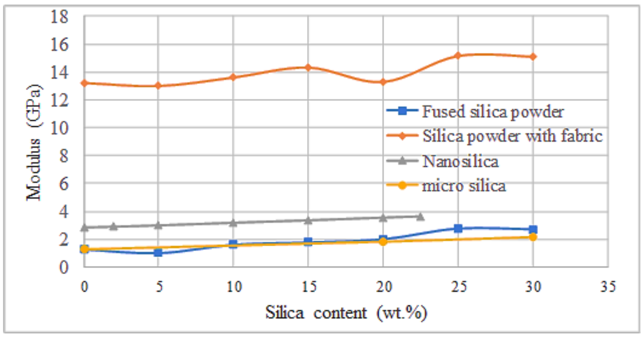

Fig. 5: Young's modulus of epoxy-SiO_2 composites, recalculated from data in [3, 18, 42].

According to Fig. 5, the ratio of nanosilica content that progressively increases implies that Young's modulus will increase as the ratio grows. Fused silica powder showed a lower increment in Young's modulus when compared to composites containing nanosilica. Additionally, this strength decreased when fused silica powder was present in amounts ranging from 5 to 20 wt.%, which may be attributed to the method of dispersing particles between the two constituents [53]. When compared to composites that do not contain fabric, composites that do contain fabric have a higher Young's modulus. On the other hand, this figure decreases when silica powder is dispersed with a percentage of fabric between 15 and 30 wt.%. Because the applied load is carried via the filler most of the time via the interface, it is attributed to the fact that the mechanical properties are affected by the particle-matrix interface as well as the filler [54, 55].

4.2.3. Tensile strength

The results of tensile strength for epoxy with silica sand nanoparticles and micro silica are shown in Fig. 6.

The tensile strength increases as the ratio of silica sand nanoparticles increases up to a certain limit, as depicted in Fig. 6, where the tensile strength achieves its maximum value at 15 wt.%, for silica sand nanoparticles and 30 wt.% for micro silica. For comparison testing of epoxy, particulate interfacial adhesion and particle loading affect mechanical properties [56, 57]. Silica nanoparticles, if not correctly

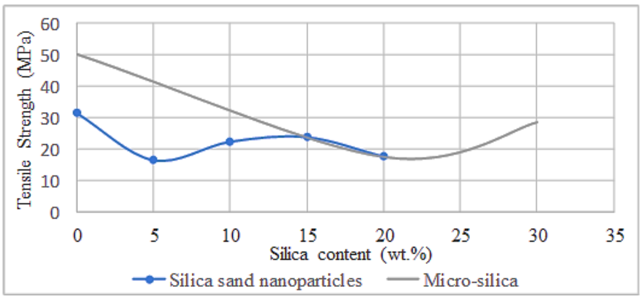

Fig. 6: Tensile strength of epoxy-SiO_2 composites, recalculated from data in Reference [36, 43].

combined with epoxy, can produce voids and increase the level of porosity in the system, leading to a lower tensile strength. This is why pure epoxy demonstrated the highest tensile strength [58–61].

4.3. Thermal properties

The thermal analysis results of epoxy and epoxy–silica nano/micro-composites are depicted in Fig. 7. (Produced from data in [35, 49].

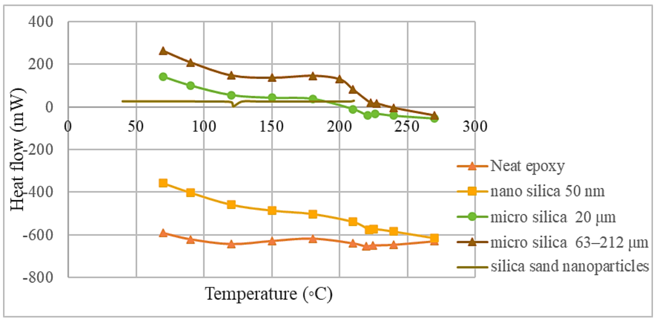

Fig. 7: Thermal study of epoxy-silica nano/micro composites, recalculated from data in [37, 49].

Differential scanning as shown in Fig. 7, calorimetric investigations of epoxy/silica systems revealed a rising trend for the glass transition temperature. Where neat epoxy exhibited glass transition temperatures between 219.45 °C. At 222 °C, the values improve slightly when 3% nanosilica is added to epoxy. Epoxy with 20% silica sand nanoparticles had a temperature of 121.59 °C. The decrease in the degree of the glass transition temperature was primarily attributable to the decreasing trend of the heat of fusion from 3 wt.% to 20 wt.% of nanosilica in epoxy, the crystalline temperature rises as the weight proportion of silica particles in epoxy increases [37]. The glass transition temperature of nanosilica including pores of 50 nm increased to 223 °C. While nonporous micro silica of 20 μm showed a smaller increase in glass transition temperature to reach 220.7 °C. This indicated the integration of a lower density of polymeric chain into the nonporous silica fillers, attributable to the lower cross-linking density of the nonporous silica micro composites [49].

5. Conclusions

the most important conclusions that can be drawn from the present study are as follows:

1. The density increases of the epoxy/silica composites with increasing silica content, with and without fabric.

2. The addition of silica fume raises the hardness of epoxy/silica composites up to a specific weight percent, after which the hardness declines. The increase in hardness that results from the combination of silica fume and carbon fiber is fixed.

3. Almost linearly, the modulus increases as the level of silica nanoparticle addition increases. While data demonstrated an increase in modulus with increasing silica powder concentration, the relationship was not always linear.

4. Tensile strength was observed to increase until a certain limited wt.% nanoparticles of silica sand in the case of polymeric composites. Data showed a lack of adequate mixing of the nanoparticles results in decreased mechanical characteristics of the nanocomposites that contain silica sand.

5. The outcomes show that the incorporation of microsilica enhanced the thermal stability of epoxy resins higher than nanosilica, attributable to the higher cross-linking density than nanocomposites of nanosilica as well as the rise in silica weight percentage.

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