

Article

The Influence of Copper Oxide Particle Size on the Properties of Epoxy Resin

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Abstract: This study examines the relationship between the size of copper particles and the properties of epoxy resin. Epoxy resin is a type of thermosetting resin commonly used as a matrix in polymer matrix composite materials reinforced with glass or carbon fibers. As part of this study, three microscale and two nanoscale composite samples modified with copper oxide particles of varying sizes were produced. This study included mechanical property tests such as static tensile tests, static bending tests, and impact tests. The results of the strength tests were compared to modeling results. Additionally, an accelerated thermal aging process was conducted to determine the impact of external conditions on the behavior of the produced composites. This study concluded with an analysis of thermal conductivity. The test results revealed that the size of the copper particles significantly impacted the tested properties. The composites with copper oxide particles on the nanoscale demonstrated the best results. These composites have promising applications in the automotive and aviation industries due to their strength, resistance to external factors, and increased thermal conductivity, suggesting their potential for producing materials that effectively dissipate heat.

Keywords: epoxy resin; copper particle size; polymer matrix composite materials; mechanical properties; ageing



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1. Introduction

Polymer matrix composite materials have been used in construction since the 1990s. They have achieved popularity owing to the large number of combinations of materials used for reinforcement, such as glass, fibrous polyamides, and carbon reinforcements. Their weight is four times lower than that of commonly used construction materials while maintaining similar mechanical properties. The greatest advantage of polymer composites is their corrosion resistance, which translates to their wide use in economic sectors dealing with the processing of chemically active materials. Examples of interesting applications include polymer composites, which can be found as elements supporting the vaults of drilled mining tunnels. Their main advantages include their low weight and electrical insulation. Materials of this type do not produce sparks during grinding and cutting, which is a highly desirable feature [1]. The storage of liquefied petroleum gas (LPG) typically involves the use of transportation and steel tanks, which have been increasingly replaced by composite tanks in recent years. The reason for this is the winding method used to produce tanks made of carbon prepreg. These composite tanks offer a volume-to-weight ratio that is four times higher than conventional tanks, resulting in a significant reduction in vehicle weight, increased fuel efficiency, and decreased pollution. Therefore, these tanks are widely utilized in industries that manufacture land and air vehicles, as well as in rescue operations, for water transport for firefighting helicopters, and as pressure tanks for filling rescue pontoons [2]. The utilization of polymer composites

in the Airbus A380 aircraft accounts for approximately 22% of its overall weight. These composites are typically layered, or laminate, structures that incorporate various materials. Examples of such composites include GFRP (“glass-fiber-reinforced polymer”), CFRP (“carbon-fiber-reinforced polymer”), and GLARE aluminum matrix composites (“glass-reinforced laminate”). These materials are favored due to their high stiffness relative to their low weight and are commonly employed in the construction of fuselage plating, wings, and tails. The anticipated demand for new materials for military vehicles in the near future will likely drive the innovation and development of materials utilized in civilian transportation [3,4]. Thermoplastics, including chemo- and thermosetting resins, are widely utilized as materials for polymer composite matrices, commonly serving as fibrous reinforcements. Notable representatives of these materials include unsaturated polyester and vinyl ester resins, polyamides, bismaleimides, phenyl-formaldehyde resins, and epoxy resins. Among these, epoxy resins are available in various molecular weights. The most commonly utilized epoxy resins are bisphenol resins, which are employed as matrices for glass or carbon reinforcements. These resins are produced through the polyaddition of bisphenol and glycerine epichlorohydrin [5].

Epoxy resins can be described using two key measures: the epoxy number and the hydroxyl number. These figures indicate the quantity of hydroxyl and epoxy groups present in every 100 g of the substance. A greater hydroxyl number signifies a higher viscosity or softening point, as well as a lower proportion of epoxy groups, which can affect the form of the substance. This may take the form of a viscous liquid or semi-solid [6]. The resin hardness is increased and retains its shape during the cross-linking process. The resulting oligomer chains are subjected to various cross-linking processes, most often at high temperatures. The cross-linking of the resin involves a copolymerization reaction involving the double bonds of both components. The main strength properties, including hardness, HDT deflection temperature, elastic modulus, tensile strength, and elongation at break, depend on the cross-linking process [7]. The use of epoxy resins is preferred over polyester resins due to their superior adhesion to fibers. Epoxy resins exhibit better wettability, leading to increased strength parameters and enhanced resistance to water. Additionally, after curing, epoxy resins do not contain ester groups, which significantly improves their corrosion resistance. An analysis of various studies provides pertinent information on the comparison of wettability between epoxy and polyester resins. Mohan [8] found that difunctional epoxy displays superior wettability among thermoset resins, regardless of the surface structure of the carbon nanotube material. Additionally, Nodehi [9] highlighted the improved wettability achieved by curing epoxy resin with specific surface sizing, indicating its effectiveness in fostering adhesion with carbon fibers. Furthermore, Bing et al. [10] characterized the wettability between carbon fibers and an epoxy resin matrix, emphasizing the significance of dynamic contact angle measurements in evaluating their interface. In contrast, Landowski et al. [11] and Cui et al. [12] discussed the challenges arising from poor wettability between polyester resins and materials such as thiolenic resins and natural fibers, respectively. These studies emphasized the importance of addressing wettability issues to enhance the performance of composite materials. Overall, the literature suggests that epoxy resin generally exhibits superior wettability characteristics compared to polyester resin, attributable to factors such as resin structure, curing processes, and surface treatments. It is essential to understand and optimize wettability to achieve robust interfacial bonding and enhance the overall performance of composite materials. These advantages make epoxy resins suitable for replacing polyester resins in the production of prepregs, which are used in the aviation industry. Table 1 provides a comparison of the mechanical properties of the most commonly utilized thermosetting resins.

Epoxy resins offer several advantages when used with a hardener. They have a longer working time, which facilitates application, and exhibit low shrinkage during the hardening process. Additionally, they can be hardened at temperatures as low as 7 °C, which makes them suitable for use in colder environments. However, they come with some drawbacks, including their higher price, which is typically double that of polyester resin. Moreover,

the amine hardeners used with epoxy resins can be toxic, and the need to use thinners and plasticizers increases the overall cost [9].

Table 1. List of selected properties of thermosetting resins. Adapted with permission from Ref. [8]. 2013, Taylor & Francis Online and from Ref [9]. 2022. Springer.

Properties	Type of Resin		
	Polyester	Vinylester	Epoxy
Density, g/cm ³	1.23	1.04	1.14
Tensile strength, MPa	70	85	75
Modulus of elasticity, GPa	3.8	3.3	3
Elongation at break, %	2.3	5	5
Thermal expansion coefficient, 6–10/°C	80–160	65	80–120

Epoxy composites also incorporate reinforcement in the form of particles, such as silica particles. These particles may be used individually or in conjunction with a fibrous reinforcement to enhance the strength and durability of the material [10–13].

Singh et al. examined the influence of the size and volume fraction of spherical silica particles on the fracture toughness of epoxy composites. The researchers incorporated fractions ranging from 1.56 µm to 240 nm and volume fractions from 0 to 0.35 into the epoxy resin. The study revealed that the fracture toughness significantly improved with an increase in volume fraction and a decrease in diameter. It was observed that as the volume fraction of small particles increased, so too did the fracture toughness. Furthermore, the researchers noted that the size of the particles played a role in the enhancement of strength when smaller particles were utilized [14]. Ahmad et al. conducted a study comparing the properties of mineral silica-based composites to conventional fused silica composites, which are commonly used as filler materials. The study examined the effect of three different particle shapes—angular, cubic, and elongated—on tensile strength, modulus, flexural strength, modulus, thermal stability, and Tg at filler contents ranging from 15 to 45 wt%. The results showed that the addition of particulate filler increased the mechanical properties of the composites while also reducing the coefficient of thermal expansion. The elongated silica mineral particles demonstrated higher flexural strength and tensile properties, as well as a comparable coefficient of thermal expansion, compared to composites with fused silica-based fillers, features attributed to the higher aspect ratio of the elongated particles and thus a stronger bond between the resin and the filler [15].

In their work, Sanya et al. examined the influence of the particle size and content of silicon carbide particles in an epoxy matrix on the mechanical properties of epoxy composites. The study investigated the effects of particle sizes of <45 µm and 90–45 µm and tested densities of 2, 4, 6, and 8 wt%. SiC. The results revealed a strong negative correlation between hardness and flexural strength, as well as a positive correlation between hardness and the flexural modulus and between compressive stress and the compressive modulus. The test results also indicated that the addition of smaller silicon carbide particles (<45 µm) resulted in excellent mechanical properties in terms of bending and compression and hardness test results. Furthermore, it was observed that an increase in SiC particle content led to a decrease in flexural strength, while compressive strength and hardness increased with increasing particle loading [16]. Abenojar et al. investigated the influence of extremely small SiC particles on the nano- and micro-scales, using particle contents of 6% and 12%. The results showed that the incorporation of silicon carbide decreased the glass transition temperature Tg due to a reduction in cross-linking points. This effect was less pronounced when using nanoscale particles. The mechanical test results revealed heterogeneity attributed to the non-uniform distribution and agglomeration of nanoparticles in the composite, as well as problems with wettability leading to material porosity. However, it was clearly demonstrated that nanoparticles increased the material's plasticity when

bending. Additionally, 6% nano-SiC significantly reduced die wear. Microparticles were found to be an effective additive for increasing the strength of the composite, but they decreased wear resistance [17].

Jayaseelan et al. investigated the influence of fiber reinforcement and macro- and microparticles of banana on the characteristics of epoxy resin, focusing on the properties of tensile strength, bending strength, impact strength, and hardness. The study involved testing composite materials with different additive contents ranging from 25 to 35% by weight. The outcomes of the tests revealed that while fibers contributed to the highest tensile strength, the particles led to the greatest increase in the bending modulus. Additionally, both banana fibers and macroparticles were found to enhance impact strength compared to unmodified resin. Furthermore, the mechanical properties of the composites were observed to improve as the additive content of both fibers and particles increased [18]. In his study, Asi analyzed the impact of Al_2O_3 particles on the characteristics of epoxy–glass composites. The Al_2O_3 content in the composites was between 2% and 15% by weight. The findings revealed that the tensile strength and shear strength of the composites significantly decreased as the Al_2O_3 content increased. However, the addition of aluminum oxide particles resulted in an increase in Young's modulus and bending strength with a rise in particle content, up to 10% by weight, and then a decrease [19]. Li et al. conducted a study to examine the influence of aluminum oxide particle size on the dielectric and mechanical properties of epoxy composites. The study's findings demonstrated that incorporating nanofillers can enhance the breakdown strength and discharge resistance of the dielectric. Conversely, microfillers can result in a decrease in short-term puncture strength with an increase in content, a phenomenon attributed to the emergence of more defects. However, it was observed that the short-term puncture strength of the nanocomposite increased with an increase in the level of nanofiller dispersion [20]. Zhou and Yu conducted research to investigate the impact of aluminum oxide on the thermal and dielectric properties of epoxy composites. Specifically, they examined the effect of varying the microparticle content at 4%, 9%, 14%, 21%, 28%, 37%, and 48% volume. The study revealed that the incorporation of aluminum particles had no effect on the thermal stability of the epoxy composites. However, it did reduce the glass transition temperature (T_g). The size, concentration, and surface area of the aluminum particles were found to influence the thermal conductivity and dielectric properties of the composites. The dielectric permittivity increased with an increase in the aluminum particle content. Furthermore, the dielectric permittivity and losses increased with temperature due to the segmental mobility of the polymer molecule [21].

Copper oxide, also known as copper(II) oxide, is a semiconductor compound with a monoclinic structure that has attracted considerable interest due to its status as the simplest member of the copper family and its potential for various physical properties. These properties include high-temperature superconductivity, electron correlation effects, and spin dynamics. CuO is an essential p-type semiconductor with a wide range of applications such as in gas sensors, catalysis, batteries, high-temperature superconductors, solar energy conversion, and field emission emitters. In the realm of energy saving, energy transfer fluids enriched with CuO nanoparticles can improve fluid viscosity and increase thermal conductivity. The narrow band gap of CuO crystal structures endows them with useful photocatalytic, photovoltaic, and photoconductive properties. CuO is also a more economical alternative to silver, miscible with polymers, and relatively stable in terms of chemical and physical properties. Additionally, metal oxide nanoparticles with a high degree of ionization, such as CuO , can be useful as antimicrobial agents due to the possibility of producing them with an extremely large surface area and unusual crystal morphology [22]. Copper oxide nanoparticles are typically available in the form of a brown-black powder. They can be reduced to metallic copper under the influence of hydrogen or carbon monoxide at elevated temperatures. These nanoparticles have diverse applications in fields such as catalysis, gas sensors, magnetic storage media, batteries, solar energy transformers, semiconductors, and field emission. Copper oxide nanoparticles, with their P-type semiconductor properties and narrow band gap, have garnered significant interest for their potential use in elec-

tronic, optoelectronic, and sensing nanostructures. They are commonly utilized as effective heterogeneous catalysts, characterized by their high activity and selectivity in oxidation and reduction reactions [23]. Copper oxide also exhibits bactericidal properties, making it suitable for use in food-processing equipment as a means of self-sterilizing the environment and preventing the spread of microorganisms. The increasing cost of silver particles has led to a shift toward using copper as a more cost-effective alternative. Copper's ability to generate toxic hydroxyl radicals, which can disrupt bacterial cell membranes, contributes to its effectiveness in combating microbial growth [24].

The primary objective of this investigation was to explore the viability of manufacturing and employing epoxy-resin-based composites that are modified with copper oxide particles. The variable component in the examined materials was the size of the reinforcement particles, ranging from 10 nm to 10 μ m. As part of the evaluation process, basic physical and mechanical properties were determined using assessments such as impact tests, and strength properties were assessed through static tensile tests and static bending tests. Accelerated aging tests were conducted to determine the influence of environmental conditions on the properties of the composites. Additionally, scanning electron microscopy was used to capture microscopic images of the materials produced, and the thermal conductivity of the composites was measured.

2. Materials and Methods

2.1. Materials

The test samples were made using LG206 epoxy resin and HG359 hardener (GRM Systems, Bielsko-Biała, Poland), and copper oxide particles of various sizes were used as reinforcements, that is, in the ranges of 1–10 μ m, 40–60 nm, and 10–30 nm, (Suzhou Canfuo Nanotechnology Co., Ltd., Suzhou, China). The resin-to-hardener weight ratio was 100:34. The reinforcement content corresponded to 10% of the weight. The sample production process included accurately weighing the proportions of resin and hardener on a Radwag 22 W laboratory scale (Radom, Poland), the ingredients were then mixed using a SH-II-7C series mechanical digital mixer up to 40 L, with a power of 80 watts (Chemland, Stargard, Poland). Mixing was carried out at a speed of 500 rpm by gradually introducing copper oxide powder. In order to obtain a homogeneous suspension, a dispersion disk stirrer with a diameter of 5 cm was used. The mixing time lasted approximately 5 min after the introduction of the reinforcement. After mixing the ingredients, the mixture was placed in a vacuum chamber. During the production of the samples, a VC3028AG vacuum chamber (VacuumChambers, Białystok, Poland) was used to minimize porosity resulting from, among other factors, mixing the ingredients. The mixture was de-aerated at a pressure of 10 mbar (abs). The hardening process was carried out at room temperature. The setting time was 24 h, and the test samples were demolded 7 days after their production. Samples for strength tests were made in accordance with the standard for plastics PN-EN ISO 3167 [25], and type A shapes were selected at dimensions of 120 mm $\times$ 10 mm $\times$ 4 mm, whereas samples for the thermal conductivity tests had the shape of a plate with the following dimensions: 150 mm $\times$ 24 mm.

2.2. Methods

Sample density tests were performed using the hydrostatic weighing method on a Radwags 22 W hydrostatic scale (Radom, Poland). As part of the mechanical property tests, static tensile, static bending, and impact tests were conducted. The static tensile test was performed on a Shimadzu AGS-X testing machine in accordance with the PN-EN ISO 527-1:2019 standard [26]. The test speed was set to 10 mm/min. The static bending test was performed on an MTS Criterion Model 43 testing machine (MTS System Corp., Eden Prairie, MN, USA), according to the PN-EN ISO 178:2011 standard [27], at a test speed of 10 mm/min. Impact strength tests were performed using a Charpy hammer (Zwick/Roell HIT 5.5P.) with a hammer energy of 5 J.

The impact of the aging factors on the mechanical properties of the composites was determined. Accelerated aging tests were performed in a QUV ACCELERATED WEATHERING TESTER aging chamber (Q-LAB Corporation, Westlake, OH, USA), using a research method in which the aging processes of materials are simulated depending on temperature and humidity. The samples were exposed to elevated temperatures, light radiation, and moisture to determine their rate of degradation. In this study, the test lasted 336 h (14 d) and 850 h (35 d), and the thermal cycle included the parameters presented in Table 2. The samples were then subjected to static tensile tests.

Table 2. Parameters of one thermal cycle.

	Function	Intensity, W/mw/nm	Temperature, °C	Time, min
Cycle	UV radiation	1.55	60	08:00
	Shower	-	-	00:15
	Condensation	-	50	03:45

A thermal conductivity test was performed using an HFM 446 Lambda Eco-Line (NETZSC, Sleib, Germany) plate apparatus. The test consisted of placing the sample between two heat sources and examining the sample temperature in several places. Owing to this test, it is possible to determine the amount of heat absorbed by the sample and the heat conduction ability of the material.

3. Results and Discussion

3.1. Mechanical Properties

Table 3 shows a comparison of density and flexural properties for all tested materials. The highest density value was achieved by a composite reinforced with copper particles with a size of 40–60 nm. A general increase in density can be seen for all materials compared to the unreinforced material. The density values of the composites with the addition of copper oxide ranged from 1.26 to 1.30 g/cm³. The additive content for each was 10% by weight; therefore, the composites have similar density values, and the differences that appear in the results were most likely caused by the different shapes of the particles because particles with a size of 40–60 nm showed a dendritic character, which could result in a greater ability to create clusters in sample areas. Impact strength tests focus on the amount of energy needed to dynamically fracture a sample. The higher the impact strength value, the more energy the material absorbs, and the more plastic cracking occurs. The highest value was obtained for particles in the range of 10–30 nm. This effect was caused by the even dispersion of copper in the matrix, thus reducing the possibility of particle agglomeration, which directly reduces the mechanical properties of the material. For materials with reinforcements in the range of 40–60 nm, we notice a decrease in impact strength which may be caused by microcracks in the matrix structure. The decrease in impact strength is most often associated with the poor adhesion of particles to the polymer matrix, irregularity in the composite structure, the formation of microcracks, for example, during the manufacturing process related to matrix shrinkage, and the presence of inclusions or defects in the form of voids and air bubbles [28,29].

Table 3 also lists the results from the static bending test. An analysis of the results showed increases in bending strength and the bending modulus for each of the tested materials. The best bending properties were obtained when copper oxide with a particle size in the range of 40–60 nm was used. Both bending strength and the bending modulus increased by over 30% compared to the unmodified material. The remaining composite materials were also characterized by increased strength properties, but the increases were slightly smaller. The addition of copper oxide with the smallest grain size resulted in an increase in bending strength of approximately 25%; however, this material was characterized by the lowest modulus of elasticity compared to the other composites, which is most likely related to the increased plasticity of the material. Al-Turaif examined the

influence of TiO₂ nanoparticles on the mechanical properties of epoxy resin. The particle sizes were 17 nm, 50 nm, and 220 nm. The modification of the epoxy resin with a small percentage of 1% TiO₂ particles resulted in a marked increase in bending stresses. As the particle size decreases, the bending strength increases, and these changes are explained by the fact that smaller particles have better mechanical properties than larger particles and adhere better to the matrix [30]. In a study conducted by Ozsoy et al., the impact of Al₂O₃, TiO₂, and fly ash on the properties of epoxy resin was examined. The researchers observed a decline in the flexural strength of the composites as the age of the particles at the nanoscale decreased. This reduction in strength was attributed to the agglomeration of nanofillers at higher concentrations and the presence of weak adhesion between the filler and matrix materials at high microfiller contents. The increase in the elastic modulus as the filler content increased is attributed to the stiffening effect of the fillers on the polymer composite [31].

Table 3. Results for basic physical and mechanical properties.

Material	Density, g/cm ³	Impact Strength, kJ/m ²	Flexural Strength, MPa	Flexural Modulus, MPa	Deflection at Failure, mm
EP	1.157 ± 0.001	8.66 ± 1.32	63.9 ± 0.9	3747 ± 277	2.6 ± 0.1
10% micro Cu	1.265 ± 0.005	14.93 ± 2.11	72.4 ± 4.0	4391 ± 222	2.6 ± 0.1
10% 40–60nmCu	1.301 ± 0.005	12.10 ± 1.51	83.4 ± 9.3	4993 ± 658	2.6 ± 0.7
10% 10–30nmCu	1.267 ± 0.002	16.86 ± 3.82	79.8 ± 3.9	4245 ± 265	3.0 ± 0.1

Research by Nur and others focused on analyzing the impact of recycled copper particles. Their goal was to investigate how different particle sizes affect mechanical properties. The particle sizes were 10 µm and 300–400 µm, and the effects of mixing different particle sizes were studied. The results of the study showed that epoxy composites with a blend of recycled copper particles had higher flexural modulus values compared to composites whose filler consisted of recycled copper with a particle size of 10 µm. Smaller filler particles contributed to the creation of a more compact structure, which in turn provided the composites with better strength. The presence of particles caused local changes in the stress field which could partially reduce the effects of cavitation deformation and instead initiate plastic deformation in the surrounding matrix during shear. Smaller particles have been shown to be more desirable due to their ability to provide better bonding; however, particles that are too small may require more polymers, resulting in reduced flexural strength. On the other hand, particles that are too large can act as discontinuities, which also reduces flexural strength. Differences between particle size groups are likely due to differences in packing density. A denser fill leads to less porosity of the composite, which in turn increases flexural strength. The results suggest that the fine filler in the mixture filled the gaps between the larger particle sizes, which contributed to the increased stiffness of the composites. The use of two different particle sizes allowed for a more even distribution of particle sizes, which in turn reduced agglomeration and increased the composite's resistance to cracking [32].

The presented test results and examples of other results available in the literature confirm the influence of particle size on bending properties while also showing that this influence is not clear and depends on many factors. The research results confirm the need to conduct this type of research due to gaps in the science related to the influence of particles on the properties being tested.

A study conducted by Sharma et al. focused on the impact of food waste particle size on epoxy resin properties. The filler content was fixed at 15% by weight, and the particle size was varied, namely, (i) 100–250 mm, (ii) 350–500 mm, and (iii) 650–800 mm. The researchers investigated the effect of particle size on the impact strength of epoxy resin. It was observed that as the particle size decreased, the impact strength increased, indicating

better energy absorption during dynamic impact. Three potential reasons were proposed to explain this outcome: improved compatibility of the fine fillers with epoxy resin, reduced void content, and the finding that smaller particles do not readily detach from the matrix compared to larger particles [33].

The test results from the static tensile test are presented in Figures 1 and 2. Strength tests have shown that the introduction of copper particles has a beneficial effect on strength properties. As the particle size of the introduced additive decreased, an increasing tendency in strength properties was noticed. The best parameters were obtained for the material modified with copper oxide particles with sizes ranging from 10–30 nm. An increase in tensile strength by approximately 30% compared to the base material, a several percent increase in the elastic modulus, and an almost 40% increase in strain at break were observed. The use of larger copper oxide particles (40–60 nm) causes slight changes in tensile strength while increasing the elastic modulus. Adhesion forces between components are largely responsible for the strengthening effect in polymer composites. Adhesion is caused by forces occurring at the reinforcement/matrix interface. These forces result from the chemical structure of the matrix and the composite reinforcement in the form of Cu particles. Chemical actions can also occur due to the similarity of functional groups. Adhesion additionally depends on the degree of wetting, the magnitude of friction forces at the phase boundary, the direction of shrinkage stress, the compactness and geometry of the filler, as well as defects in the form of voids and air bubbles [34,35]. The results show that the composite with the smallest particle size has the best properties, which is probably related to the greater number of contact points at the particle–matrix interface. However, the introduction of larger particles with a dendritic structure resulted in decreases in strength and plastic properties, which may be related to the formation of agglomerations and defects in the form of voids, which weakened the composite material.

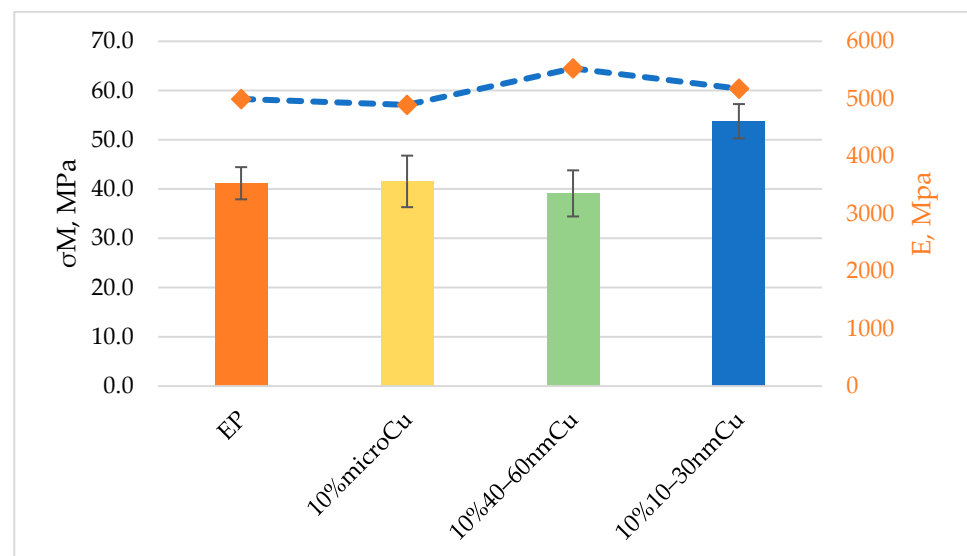


Figure 1. Comparison of tensile strength and Young's modulus.

Nourbakhsh et al. studied the effect of the particle size of 30% wood flour in a polypropylene-based composite. The test results showed that in the case of smaller particles (0.30 and 0.25 mm), there was a moderate increase in strength, and the particle size had a moderate impact on the elastic modulus of the composites. In this work, the authors also cite the results of other studies that confirm the positive effect of wood flour on the properties of polymer composites with decreasing particle size, giving possible reasons for this type of behavior. The increase in strength is attributed to better interfacial adhesion between the matrix and the particles, along with other factors influencing properties such as the particle size and size distribution of the wood, the aspect ratio and abrasion during processing, the adhesion of fibers to the matrix, the transfer of stresses at the interface, and

temperatures mixing [36]. Sharma et al. reached similar conclusions. They investigated the influence of the size of food waste particles on the properties of epoxy resin. The filler content was 15% by weight. The particle size was (i) (100–250) μm , (ii) (350–500) μm , and (iii) (650–800) μm . The tests showed a decrease in mechanical properties with an increase in particle size. These results were explained by the fact that larger particles detach more easily than small particles. This debonding results in more voids, which ultimately reduces the stiffness of the composites [33]. In turn, Ozsoy and others pointed out other potential problems related to the introduction of micro- and nanoscale additives. They examined the influence of Al_2O_3 , TiO_2 , and fly ash on the properties of epoxy resin. The filler content ranged from 10 to 30 wt%, and the particle sizes were 10 nm and 40 nm and 45 and 50 μm . The results clearly show that the tensile strength of microfiller composites decreases with increasing filler content. This can be explained by the fact that increasing the filler content caused poor adhesion between the matrix and the fillers and led to a decrease in the strength of the epoxy composite. In the case of the nanofiller, the decrease in strength was due to the uneven distribution of fillers at high filler proportions, which led to agglomeration and caused areas of stress concentration, which led to some decrease in strength [31].

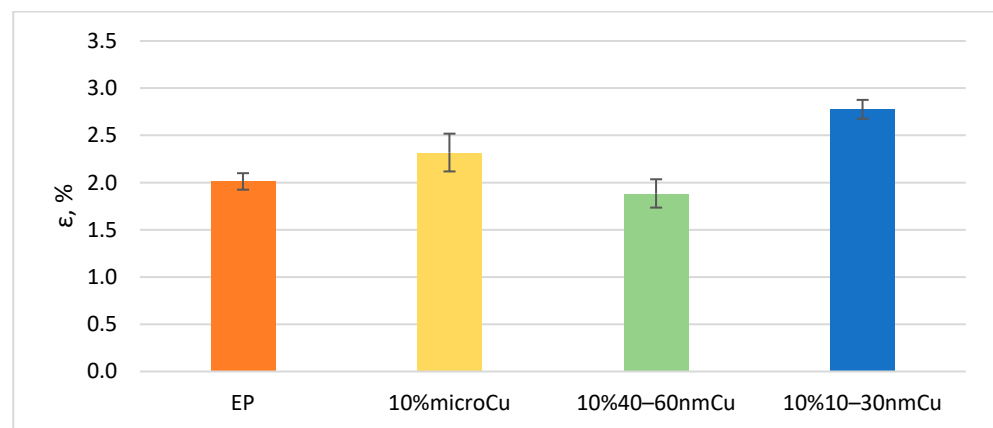


Figure 2. Comparison of strain at break values.

Fu and others presented in their work a summary of various studies on polymer matrix composites modified with particles of different sizes. Conclusions can be drawn from their work. In summary, there appears to be a certain critical particle size beyond which no significant effect on the modulus of the composite is observed. When the particle size is smaller than this critical value, the particles' effect on the modulus of the composite becomes more significant. However, the precise value of this critical particle size cannot be predicted in advance because it depends on various factors such as the properties of the particle, the matrix, and their mutual adhesion. This work presents a comprehensive review of experimental results and theories regarding the mechanical properties, including the modulus, strength, and fracture toughness, of micro- and nanocomposites containing polymer-based particles. The influence of particle size, particle adhesion to the matrix, and particle loading on composite stiffness, strength, and crack resistance was analyzed in detail, examining a wide range of composites that contain both micro- and nanofillers of various shapes. It has been shown that all three factors mentioned have a significant impact on the strength and resistance of the composite, especially the adhesion of particles to the matrix. This is expected because the effective stress transfer between the filler and matrix and strength/brittleness are strongly related to adhesion. Various trends in the influence of particle loading on the strength and resistance of the composite were observed which result from complex interactions between the factors studied. However, the stiffness of the composite depends largely on particle loading and not only on particle–matrix adhesion because fillers usually have a much higher modulus of elasticity than the matrix. There is also a critical particle size value, usually on the

nanometer scale, below which the stiffness of the composite increases significantly, probably due to the increased surface influence of “nano” effects [37].

3.2. Mechanical Modeling

Particle size, particle–matrix adhesion, and particle loading stand out as important determinants in understanding the behavior of a composite material. Several parameters are intricately linked to determine strength and mechanical characteristics such that the quality of particle adhesion to the matrix has a major effect on a composite’s strength and toughness. It is observable that when particle sizes are in the micron or greater range, the Young’s modulus values of composites are insensitive to their size. On the other hand, as particle sizes approach the nanoscale, a notable increase in stiffness is shown [38]. It was shown that micro-fibrils with sizes between 10 and 30 nm have been seen to influence interphase boundaries more significantly than those in bulk. The composite modulus is expected to increase with decreasing particle size, with a critical particle size of 30 nm [37]. It is generally known that the smaller the dispersed phase, the greater its specific surface area, resulting in shorter distances within the vicinity of the dispersed phase when the volume fraction of the dispersed phase remains the same. This also means that the mechanical responses of a thin layer placed between the stiff dispersed phase and the matrix would differ from those of the dispersed phase.

In this study, the effect of particle size on the Young’s modulus of polymer composites is shown. To describe this effect, a three-phase Takayanagi model [39] was used which provides better insight into the intricate interaction of various components. The averaged Young modulus of composite E_c was calculated by following formula:

$$\frac{1}{E_c} = \frac{1 - \sqrt{[(r + \tau)/r]^3 \phi}}{E_m} + \frac{\sqrt{[(r + \tau)/r]^3 \phi} - \sqrt{\phi}}{\left\{1 - \sqrt{[(r + \tau)/r]^3 \phi}\right\} E_m + \sqrt{[(r + \tau)/r]^3 \phi} (\kappa - 1) E_m / \ln \kappa} + \frac{\sqrt{\phi}}{\left\{1 - \sqrt{[(r + \tau)/r]^3 \phi}\right\} E_m + \left\{\sqrt{[(r + \tau)/r]^3 \phi} - \sqrt{\phi}\right\} (\kappa + 1) E_m / 2 + \sqrt{\phi} E_r} \quad (1)$$

where E_m , E_r are, respectively, the Young’s modulus values of the matrix and reinforcement, r is the radius of a spherical particle, τ is the thickness of the interfacial region, and κ denotes a linear gradient change in the modulus between the matrix and the particle surface, indicating the modulus ratio of the adjacent interface to the particle surface. In Equation (1), the term ϕ denotes the volume fraction of the second phase instead of the mass proportion *wt*. Thus, this fraction should be calculated using the following equation:

$$\phi = \frac{1}{1 + \frac{\rho_f}{\rho_m} \left(\frac{1}{wt} - 1 \right)} \quad (2)$$

where ρ_f and ρ_m represent, respectively, the density of the reinforcing phase and the matrix density.

Additionally, the Voigt and Reuss models were used as theoretical frameworks for comparisons with experimental data, aiming to understand the elastic properties of a composite with different particle sizes. The Voigt model [40], based on an assumption of uniform strain, provided an upper bound estimate of the stiffness of the material by averaging the stiffness tensors.

$$\bar{E}^V = \sum_{j=0}^k \phi^j E^j \quad (3)$$

where ϕ^i is the volume fraction of i -th phase, labelled $i = 0, \dots, k$. Conversely, the Reuss model [41], which assumes uniform stress, provides a lower bound.

$$\bar{E}^R = \left[\sum_{i=0}^k \phi^i (E^i)^{-1} \right]^{-1} \quad (4)$$

The calculations were based on the properties described in this paper with an exception in which 70.5 GPa was taken as the copper oxide stiffness based on the work of Lehmann et al. [42]. The density of CuO was 6.31 g/cm³. The value of the volume fraction obtained for the adopted data was 0.02. The values of stiffness for nanoscale particles inside the area were described by theoretical models (Figure 3a). However, for the composite in which larger particles were used, a lower Young's modulus value was characterized than in the presented estimations.

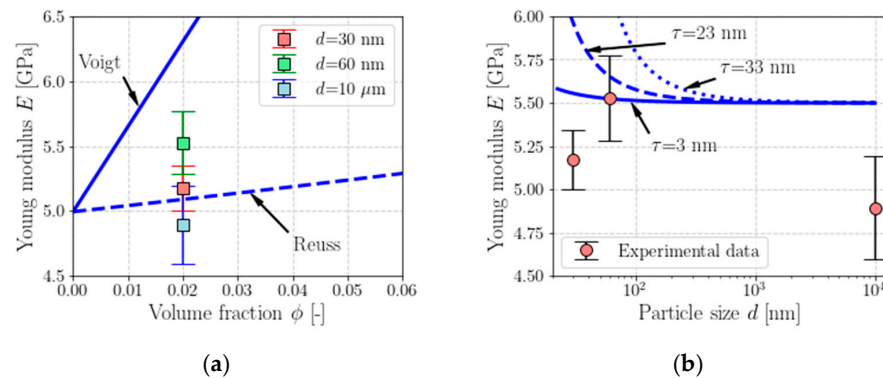


Figure 3. Comparison of experimental results for different particle sizes with theoretical evaluations using the Voigt and Reuss models (a) and estimations of the composite Young's modulus based on Equation (1), where $\kappa = 4$ (b).

It is important to note that the Voigt and Reuss models are generally used to estimate the bounds of the elastic properties of bulk materials. They do not take into account the influence of particle size or other microstructural characteristics that can significantly affect the mechanical properties of materials. Their role was to provide theoretical boundaries for the elastic moduli of the materials. The deviations observed in the experimental data, related to changes in particle size, highlight the limitations of these models in capturing the effects of microstructural variations.

Analytical estimations using the Takayanagi model exhibited higher stiffness values than the experimental data (Figure 3b). The increase in the thickness of the interfacial region results in stiffer behavior of the material. However, in the micro-size range, this effect is negligible. The level of the approximated Young modulus is close to results of the Voigt model but, as previously mentioned, this simple model does not consider the size of the reinforcement particles. This is the reason why the Voigt and Reuss models cannot be directly compared with the Takayanagi model. Nevertheless, the three-phase model does not accurately describe the empirical data, which leads to the conclusion that it requires further development.

3.3. Microscopic Observations

As part of this work, an analysis of the microstructure of the base material and the produced composites was carried out. Figure 4a,b show the structure of epoxy resin. This material is characterized by typical brittle fractures. Figure 4c–h present microscopic photographs of both the copper oxides used and the composites modified with them. The copper oxides with particle sizes of 1–10 μ m and 10–30 nm show similar types of fine-grained particles which are characterized by a whisker-like structure, while the copper oxide particles with a size of 40–60 nm are characterized by a more developed, dendritic structure. An analysis of the photographs of the microstructures of the produced composites showed that composites with copper with particle sizes of both 1–10 μ m and 40–60 nm are brittle, while the use of copper oxide with a particle size of 10–30 nm changes the

nature of the matrix to a more developed one, which suggests some plasticization of the material. Additionally, strength tests seem to confirm this effect because the materials were characterized by the highest deformation at break and the highest impact strength value.

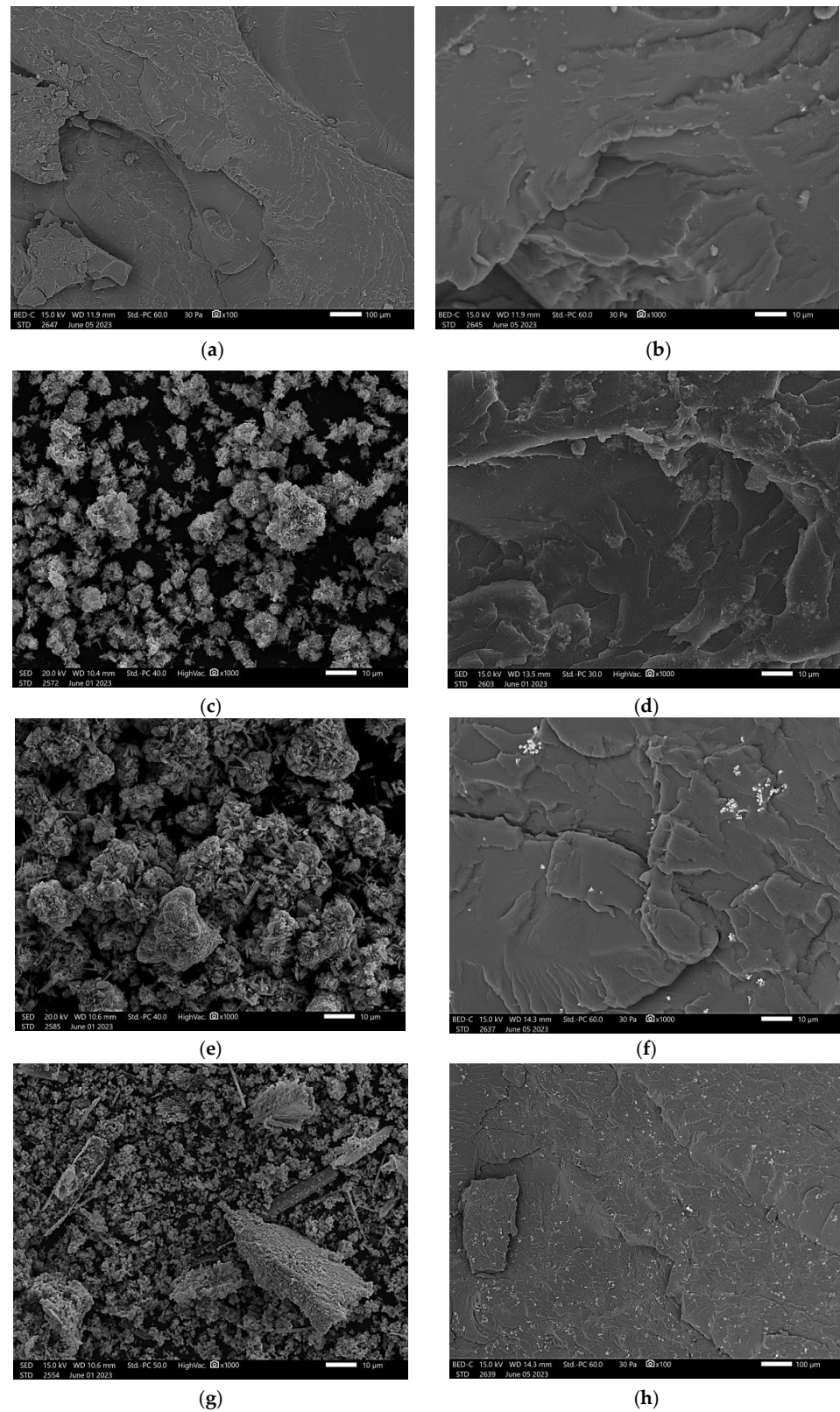


Figure 4. Structure of copper oxide powders and manufactured composites. (a) Structure of epoxy resin fracture. (b) Structure of fracture structure of epoxy resin. (c) Structure of copper particles with

a size of 1–10 μm . (d) Fracture structure of resin composite reinforced with copper particles with a size of 1–10 μm . (e) Structure of copper particles with a size of 40–60 nm. (f) Fracture structure of resin composite reinforced with copper particles with a size of 40–60 nm. (g) Structure of copper particles with a size of 10–30 nm. (h) Fracture structure of resin composite reinforced with copper particles with a size of 10–30 nm.

3.4. Accelerated Aging

The manufactured materials were also subjected to accelerated aging tests. Aging is a picture of all the irreversible physical and chemical changes occurring in a material at a specific time. The tested conditions were temperature, chemical compounds, and mechanical loads. When all these parameters are known, it is worth increasing one factor to speed up the test.

We mention two types of aging: internal and external. The internal aging of polymer materials is the result of thermodynamic instability, which manifests itself through recrystallization, the relaxation of residual stresses, the dispersion of phases in multi-component systems, and the migration of plasticizers. On the other hand, external aging refers to processes such as thermo-oxidative degradation, the formation of stress and fatigue cracks, and swelling, which are directly related to environmental chemical or physical impacts on plastic. Aging is also divided into physical and chemical aging, but a given environmental factor can cause both effects at the same time, which significantly complicates the clear affiliation to a given group, e.g., increased temperature can cause both physical (secondary crystallization) and chemical (chain breakdown) effects. Changes in structure, secondary crystallization, and disorientation or the relaxation of residual stresses are physical phenomena typical of aging processes. Physical aging phenomena may occur during the evaporation or migration of volatile components or with mass increases through water absorption or swelling. However, the chemical structure is most often changed under the influence of chemical effects such as the impact of heat in combination with oxygen or other aggressive substances or radiation, with a strong emphasis on solar UV radiation. Increased temperature and, consequently, heat absorbed by the material, can initiate cross-linking, which changes the properties of the material while improving its thermal, mechanical, and physical properties. In the medium term, however, this may contribute to the degradation of the chains and the formation of cross-linking, which has a negative impact on the material. Oxygen has the ability to attach to other polymer chains, resulting in the formation of thermally unstable hydroxides, causing chain breakage. Ozone and radiation support oxidation, which accelerates the breakdown of chains. UV radiation causes the material to crumble, and in the case of condensation polymers, water combined with heat leads to the hydrolysis of the material; an example is polyamide. Four types of chemical degradation can be distinguished: (1) depolymerization, (2) chain breakdown, (3) thermo-oxidation, and (4) radiation. The first one involves the formation of monomers, while the second one can occur at any point in the chain. The last two types are related to cross-linking reactions and the detachment of substituents and their elimination. The type of degradation and the degree of changes during aging depend on the chemical structure of the material, the environmental conditions, wall thickness, and the test sample. The sample's electrical, mechanical, and thermal properties are changed under the influence of the test. In addition, surface changes are noticeable, such as a change in the color of crystallinity or structure [43,44]. Figures 5–7 show test results from before and after the aging process. The applied aging conditions resulted in a significant decrease in the strength properties of the unmodified material. The introduction of copper oxide particles resulted in an increase in the stability of the tested materials, both in tensile strength and Young's modulus. The strain at break decreased with an increase in the accelerated thermal aging time for all materials, but in this case, copper particles also limited the negative impact of the external environment on the tested materials. The best results were achieved by materials modified with copper nanoparticles. In their research, Abenojar and colleagues

investigated the impact of temperature, humidity, and their combined effect on the thermal and mechanical properties of epoxy micro- and nanocomposites containing SiC. The study results indicated that under the aging conditions analyzed, water absorption and desorption exhibited a close correlation with the mechanical properties and the softening transition temperature (T_g) of the material. Specifically, water absorption led to the plasticization of the material, which in turn reduced its strength and T_g . Conversely, water desorption resulted in increased brittleness and cross-linking in the material, leading to increased strength and T_g . Moreover, an increase in temperature favored the cross-linking process of the material, contributing to improvements in the mechanical properties of the polymer composite and an increase in T_g . Interestingly, the size of SiC particle showed no significant effect on property trends, although all major changes were observed for nanocomposites compared to microcomposites [45]. It seems that in the presented research results, changing cycles of moisture and temperature caused an increase in strength after the accelerated thermal aging process, but the influence of other factors such as particle size and shape should also be taken into account. Aging changes can be seen with the naked eye; Figure 8 shows macroscopic surface changes observed with an optical microscope. The samples were additionally characterized by a change in color and a cracked surface through which copper oxide particles were visible.

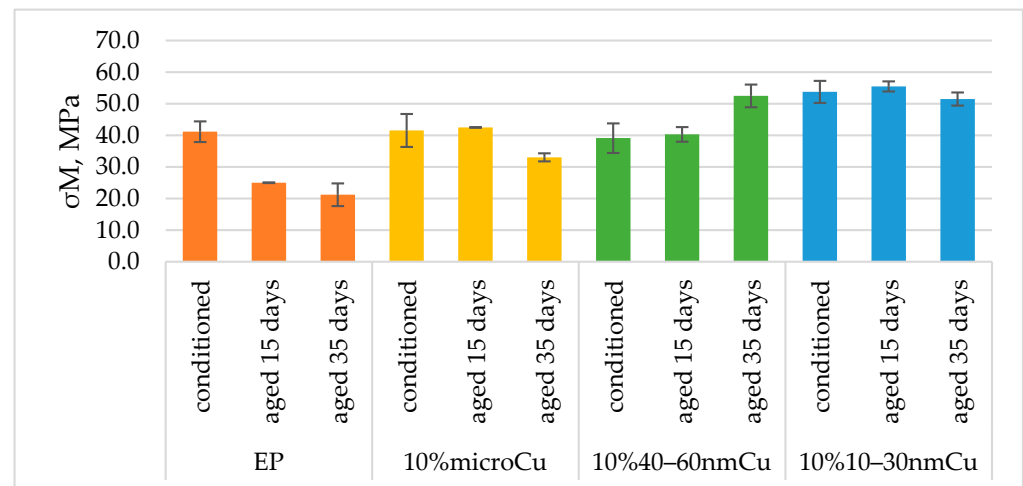


Figure 5. Comparison of tensile strength values before and after the accelerated thermal aging process.

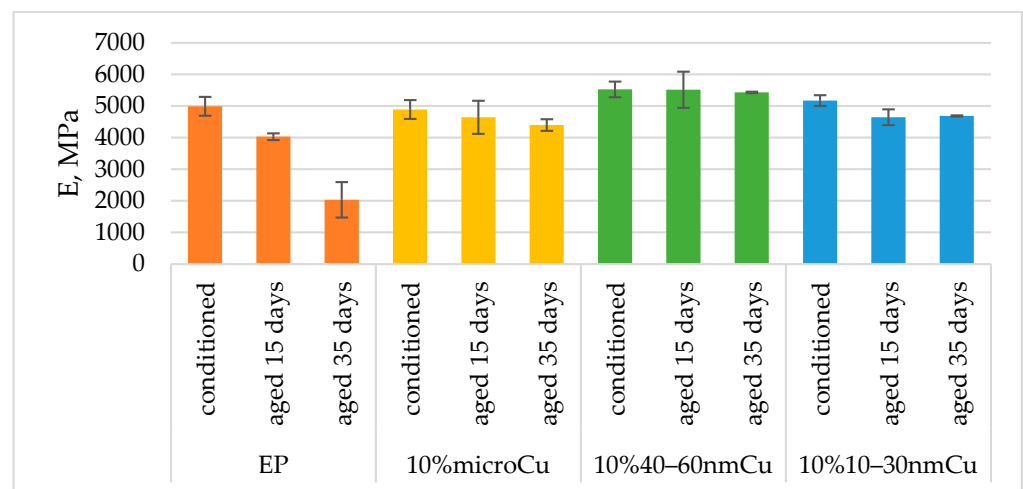


Figure 6. Comparison of Young's modulus values before and after the accelerated thermal aging process.

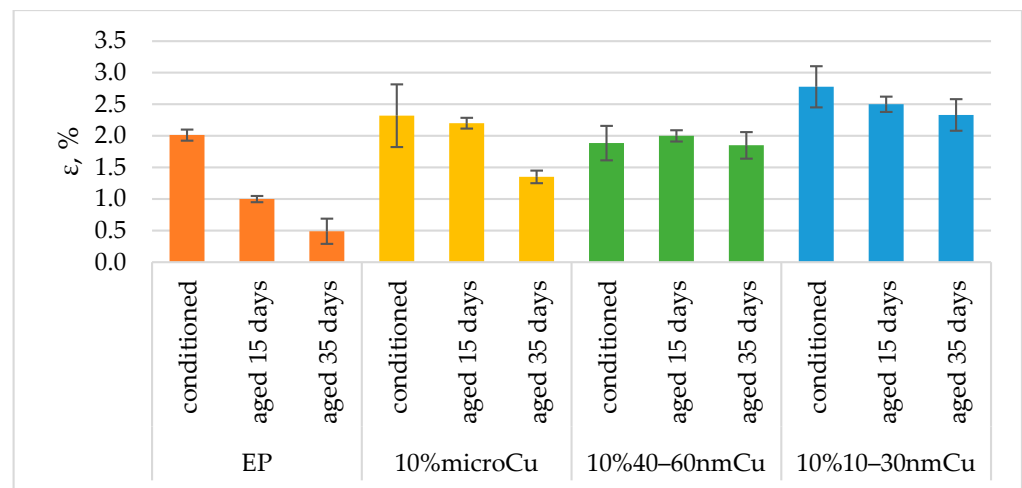


Figure 7. Comparison of strain at break values before and after the accelerated thermal aging process.

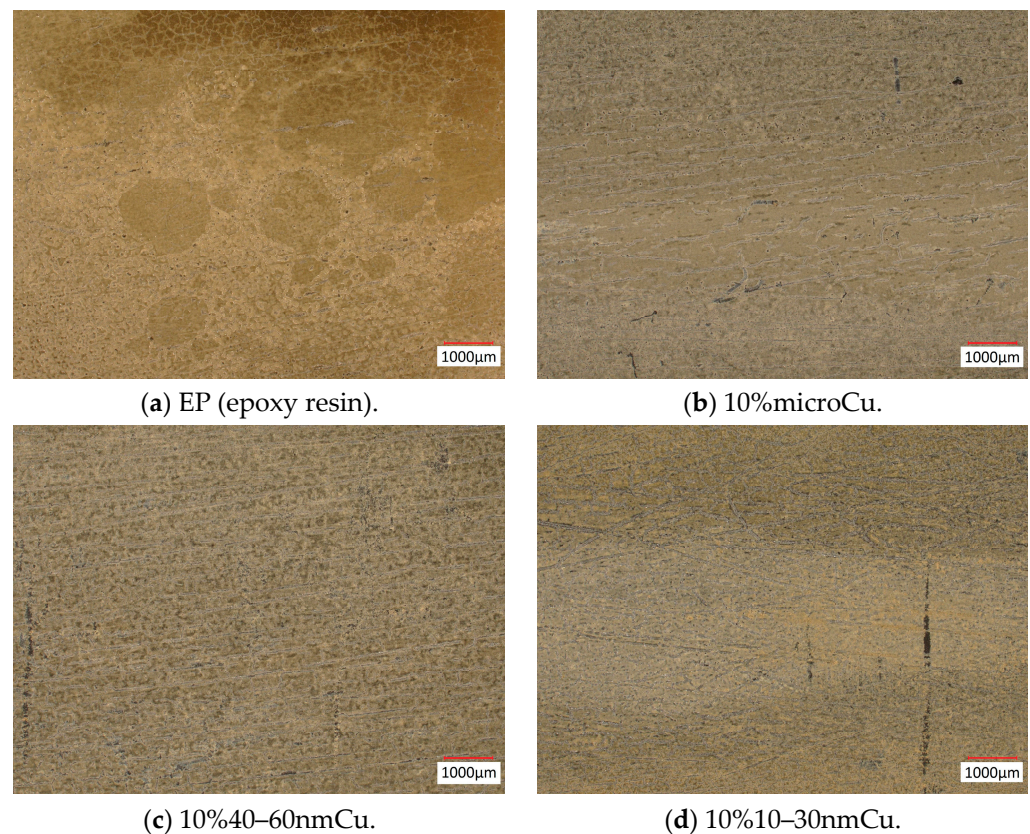


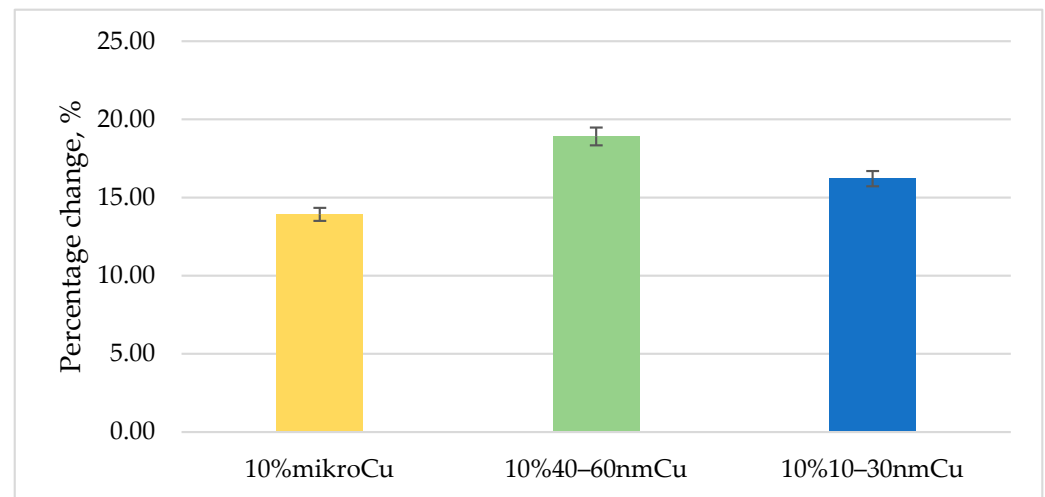
Figure 8. Macroscopic photos of sample surfaces after the accelerated thermal aging process.

3.5. Thermal Conductivity

Analyzing Table 4 and Figure 9, it can be seen that the addition of copper in the amount of 10% by weight has a positive effect on thermal conductivity. The greatest increase is noticeable for particles with a size of 40–60 nm. Taking into account the given graphs, it can be concluded that the increase is caused by the natural conductive properties of copper which, after reaching a given value, creates percolation paths, enabling thermal flow. The smaller the particles, the higher the thermal conductivity value until it reaches a critical value; for Cu particles, this is in the range of 10–30 nm. The decrease in thermal conductivity may be caused by an insufficient distribution of copper particles and, consequently, the creation of significant spaces, which prevent the creation of uninterrupted conductive paths.

Table 4. Thermal conductivity results.

Parameter	EP	10% MikroCu	10% 40–60nmCu	10% 10–30nmCu
Thermal conductivity, W/(m·K)	0.21717 ± 0.007	0.24741 ± 0.007	0.25824 ± 0.008	0.25238 ± 0.008
Average temperature, K	10.5 ± 0.03	10.6 ± 0.03	10.7 ± 0.03	10.7 ± 0.03
Percentage change, %		13.92 ± 0.42	18.91 ± 0.57	16.21 ± 0.49

**Figure 9.** Percentage increase in thermal conductivity compared to the initial value of thermal conductivity of the epoxy resin.

Composites reinforced with copper particles are good conductors of electricity and heat; this is possible thanks to a phenomenon called percolation. In order for a composite to become a conductor throughout the entire material, it is necessary to form continuous conductive paths. Thanks to the formation of a conductive network, it is possible to observe the free flow of electrons through the connected paths, which guarantees an increase in thermal and electrical conductivity. A percolation barrier describes the minimum concentration of reinforcement particles needed to form conductive beams. If a given value is not achieved, the particles are randomly distributed in the material, which causes disturbances in the network, reducing the flow of electrons. This phenomenon is crucial when forming composites reinforced with copper particles because, thanks to the appropriate selection of concentration, we can achieve the desired conductivity. Moreover, the phenomenon of network formation can be improved using surface treatment techniques to improve dispersion and reduce particle agglomeration in the composite matrix [39]. Misiura and colleagues investigated the mechanical properties as well as the electrical and thermal conductivity of epoxy composites modified with copper and nickel. It was found that the electrical conductivity of the composites exhibited percolation behavior at specific threshold values of 9.9% and 4.0% vol% for EP-Cu and EP-Ni composites, respectively. Using the Lichtenecker model, the theoretical thermal conductivity of the dispersed metallic phase in the composites was estimated to be 35 W/mK for Cu powder and 13 W/mK for Ni powder. The findings of the theoretical and experimental studies overlapped. Unlike electrical conductivity, thermal conductivity did not exhibit percolation properties. These values are nearly an order of magnitude lower than the thermal conductivity of solid metals. This phenomenon can be attributed to the presence of high thermal resistances at the particle-to-particle and particle–polymer interfaces which impede heat transfer in the metal-filled polymer matrix [46].

4. Conclusions

Summing up the obtained test results, a positive effect of using copper particles as reinforcements in the context of strength parameters was noticed. An increase in mechanical properties was observed in relation to the control sample of a pure polymer for all tested composites. In addition, an upward trend in parameters was observed with reductions in individual particle sizes. The best strength parameters were achieved for composites reinforced with particles in the size range of 10–30 nm. For these materials, the best results in the static tensile test were recorded, which is consistent with the theory in the literature on the dependence of strength on adhesion forces and a larger contact surface between the matrix and the reinforcement. In addition, it was noted that larger particles cause agglomeration, which adversely affects strength properties. In the flexural strength tests, the composite modified with particles in the 40–60 nm range had the best strength. This is related to the examined structure of copper particles which, due to their developed dendritic surfaces, showed greater plastic properties. Thus, it can be concluded that the strength parameters depend not only on the size of the particles but also on their structural nature. Composites reinforced with copper oxide do not degrade under the influence of environmental conditions in the short term. Ultimately, the copper particles improve the thermal conductivity of the composite relative to the reference material. An increase of more than 18% was obtained for the material modified with particles in the 40–60 nm range, and a 16% increase was obtained for the material reinforced with particles in the 10–30 nm range, which means that reducing the particle diameter has a positive effect on thermal conductivity; however, it occurs at a certain critical value, blocking the formation of a continuous percolating network. The novelty of the field of copper–epoxy composites has been addressed in various studies. Xu et al. [47] introduced a foamed copper–epoxy composite that showed improved wear resistance, making it a unique material for abrasion-resistance applications. Nur and colleagues [32] also observed flexural strength and modulus improvements by incorporating mixed copper particles into epoxy composites, highlighting the potential for innovative material improvements. Mykhalichko and Lavrenyuk investigated using copper(II) hexafluorosilicate-modified epoxy–amine composites for flame protection, which showed progress in increasing thermal stability through chemical interactions [48]. Biswas et al.'s study on adding copper slag filler to bamboo–epoxy composites showed improved physical and mechanical properties, suggesting the innovative development of composite materials [49]. Lavrenyuk and colleagues developed self-extinguishing copper(II)-coordinated epoxy–amine composites for pouring polymer floors, demonstrating progress in creating materials with improved properties [50]. Honda et al. demonstrated a strengthening of the epoxy–copper heterojunction by incorporating sulfur-containing polymers, suggesting new approaches to improving material interfaces [51].

In summary, the research results presented, the discussion in this article, and the examples illustrated above highlight the need for the further development of epoxy composites modified with copper and copper oxides. These comparisons indicate the continuing need for research in this field and the need to create new copper–epoxy composites that can be used in various areas, ranging from wear-resistant materials and flame protection to composites with improved mechanical and thermal properties. The research studies mentioned above jointly contribute to the development of materials engineering by introducing innovative recipes and applications of copper–epoxy composites. Epoxy composites reinforced with copper particles can be widely used in the electronics industry and in control and conductive devices manufactured for transport and maneuvering machines; however, in to further develop work on a given material, its compatibility with other types of reinforcements should be checked.

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