MECHANICAL, THERMO-MECHANICAL AND TRIBOLOGICAL PROPERTIES OF SILICON CARBIDE NANOPARTICLES FILLED EPOXY COMPOSITES

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The paper presents the study of the effect of nanometric silicon carbide (nSiC) addition into epoxy composites concerning mechanical, thermo-mechanical and tribological properties. The results show that nSiC is able to improve all studied properties of the epoxy composites, e.g. tensile and flexural strength and stiffness, heat deflection temperature and friction coefficient. The paper presents a nanofiller weight content optimization study, showing that optimum results are obtained when nanometric silicon carbide is added in 1 wt.% relative to the epoxy matrix.

Keywords: epoxy resin, nano-silicon carbide, strength, friction coefficient

1. Introduction

Epoxy resins are currently widely used in the industrial, construction and aerospace field, as adhesive or as matrix for fiber-reinforced composites [1]. The high demands of the applications these materials are used for, require a continuous improvement of their properties, especially from mechanical, thermal and service life point of view. The main experimental studies focus on maintaining the epoxy matrix low weight advantage and reduce their main disadvantages such as their brittle nature that sustains crack propagation during mechanical failure [2, 3] and low thermal resistance on extended functioning periods [4].

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One of the most common practice is to improve epoxy resin properties by introducing an additional phase consisting of liquid rubbers, thermoplastics, copolymers [1] or inorganic fillers in micro or nano scale [5-8]. Fillers enhance a variety of important properties including machinability, corrosion resistance, flame retardancy, thermal stability, abrasion resistance, electrical properties, dimensional stability etc. [9]. The suitable filler is chosen depending on the targeted application, and consequently on the specific property/properties that need to be enhanced. E.g. rubber particles are added to epoxy resin for increased fracture toughness [1, 10], while rigid nanofillers, such as silica, alumina, carbon nanotubes, nanoclays can be used for increased strength, stiffness and fracture toughness [1, 11-14].

Aerospace applications require special properties, therefore amongst the variety of inorganic fillers used with epoxy resin to form nanocomposites, ceramic nanofillers are very promising. Bulk SiC is a ceramic material with high hardness, wear resistance, high temperature stability and chemical inertness that has many applications in abrasive, metallurgy, composite and electronic industries [15, 16]. These versatile properties sustained the use of silicon carbide in micro or nanometric form to achieve different polymeric composites with improved properties. Silicon carbide in micrometric or nanometric form was used in several studies to improve different properties of thermoset matrix such as epoxy [17] or phenolic resin [18, 19], as well as thermoplastic matrix such as polyolefins [20] or polystyrene [21, 22].

There are also studies that evaluate the effect of nSiC added as nanofiller into the polymeric matrix of fiber reinforced composites [5, 17, 23-25], which show promising results of superior performance of nanomodified systems compared to the unmodified one, in terms of thermal, mechanical and tribological properties. All these literature results prove that silicon carbide is a promising material as nanofiller in composites based on a wide range of polymeric matrix.

The team's research studies focused especially on phenolic resin based composites with silicon carbide nanofiller, the conducted study was one of the few presenting the properties modification of simple phenolic resin in the presence of nSiC [26], the results showing that at an optimum weight content of 1%, mechanical, thermal and tribological properties were significantly improved. The present study focuses on the evaluation of the effect of nanometric silicon carbide on epoxy resin, regarding mechanical, thermal and tribological properties. The study represents a preliminary experimental stage of a larger research, which focuses on developing advanced composite materials for aircraft and aerospace applications with improved polymeric matrix.

2. Materials and Methods

2.1. Materials

The matrix was epoxy system L20/EPH161, purchased from R&G Faserverbundwerkstoffe GmbH. The L20 epoxy resin is diglycidyl ether of bisphenol A (DGEBA) with 1.15 g/cm^3 density, 900 mPa·s viscosity and 179 g/eq epoxy equivalent, EPH161 curing agent is 3 aminopropyl-3,5,5-trimethylcyclohexylamine and was added at 25 wt.% content relative to the resin. The nanofiller agent was β type nanometric silicon carbide purchased from Nanostructured & Amorphous Materials Inc., USA, with 97.5 % purity, 34-40 m^2/g specific surface area and 3.22 g cm^{-3} true density.

2.2. Nanocomposites synthesis

The nanocomposites synthesis followed two main steps. The first step consisted of the nanofiller powder dispersion into the liquid L20 epoxy resin. The nano silicon carbide was added in 3 weight contents relative to the epoxy resin: 0.5, 1 and 2 wt.%. The mixtures were mechanically homogenized for 2-4 min and ultrasonicated using an ultrasound probe for 15 min. The second step was the curing. The curing agent (EPH 161) was added at 25 wt.% content relative to the resin, the obtained mixtures were gently homogenized and poured into Teflon moulds that were previously treated with mould release agent. Curing took place at 25 °C temperature, 2 bar pressure for 24 hours.

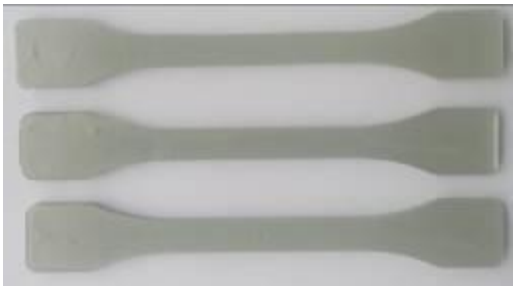

Fig. 1. Nanocomposite specimens for tensile tests

2.3. Nanocomposites characterization and testing

The nanometric silicon carbide filled epoxy materials were subjected to chemical and morphological analysis, as well as mechanical, thermo-mechanical, and tribological tests. FTIR spectroscopy analysis was performed using Spectrometer Nicolet iS50 (operated in ATR mode) and scanning electron microscopy (SEM) was performed using FEI Inspect F50 High Resolution Scanning Electron Microscope with field emission gun and 1.2 nm resolution and energy dispersive X-ray spectrometer (EDS). Mechanical tests were performed using INSTRON 5982 mechanical testing machine and consisted of 3-point bending, following SR EN ISO 178 (2 mm/min testing speed and nominal span

length- 16 x specimen thickness, on 80x10x4 mm rectangular specimens, using 5 specimens per sample) and tensile test, following SR EN ISO 527 (5 mm/min tensile rate, dog-bone shaped specimens. Heat deflection temperature (HDT) was determined using Qualitest HDT1- Heat Deflection System, following SR EN ISO 75 (2 °C/min heating rate, 1.8 MPa flexural stress, silicone oil immersion environment, 3 specimens for each sample). Friction coefficient was determined using CETR UMT 3 (Universal Macro Materials Tester) block-on ring module, on a 35 mm diameter steel role, 10 N load, for 60 seconds at two different speeds: 1000 and 1500 rpm, on 3 specimens for each sample.

3. Results and discussion

3.1. FTIR spectroscopy

FTIR analysis was performed on the grinded samples, in order to register the bulk characteristics of the materials.

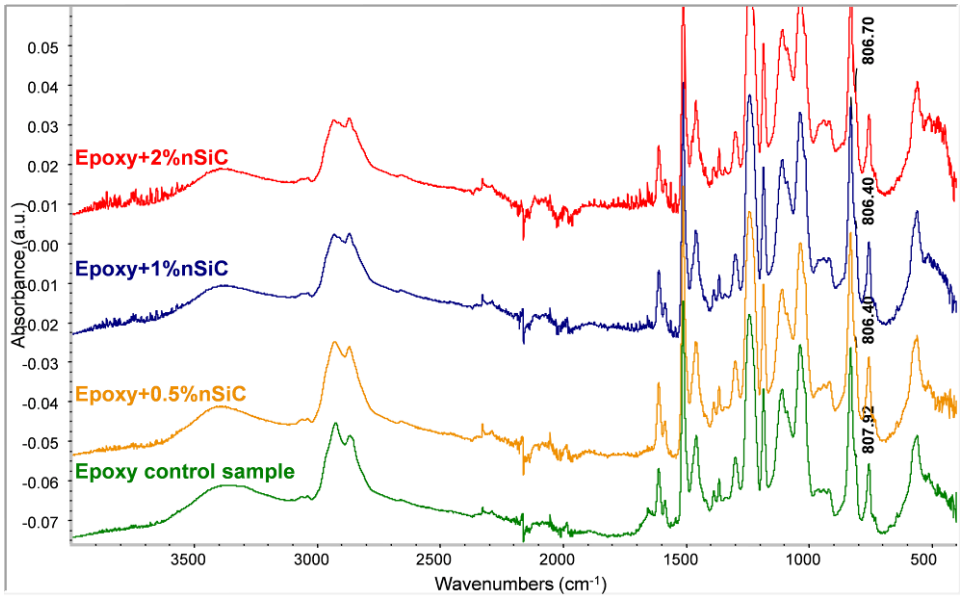

Fig. 2. FTIR spectra of nSiC filled cured epoxy composites

The control sample and nanofilled samples were subjected to FTIR spectroscopy, the nSiC filled epoxy samples spectra presenting no visible differences compared to the control sample. This could be due to a possible good embedding of the nanoparticles into the epoxy matrix, the analysis not being able to detect the specific peak of the nSiC (at 800-815 cm^{-1} [27, 28] owned to the Si-C vibration), as the nanopowder is added in very low weight contents that do not form high agglomeration areas. This result will be supplemented by SEM and EDS analysis below.

3.2. SEM analysis

Scanning electron microscopy was performed in the fracture cross-section resulted after mechanical tests (Fig. 3). The SEM images show that the nSiC nanoparticles presence influences the fracture area, but both control sample and nanofilled samples exhibit a brittle fracture.

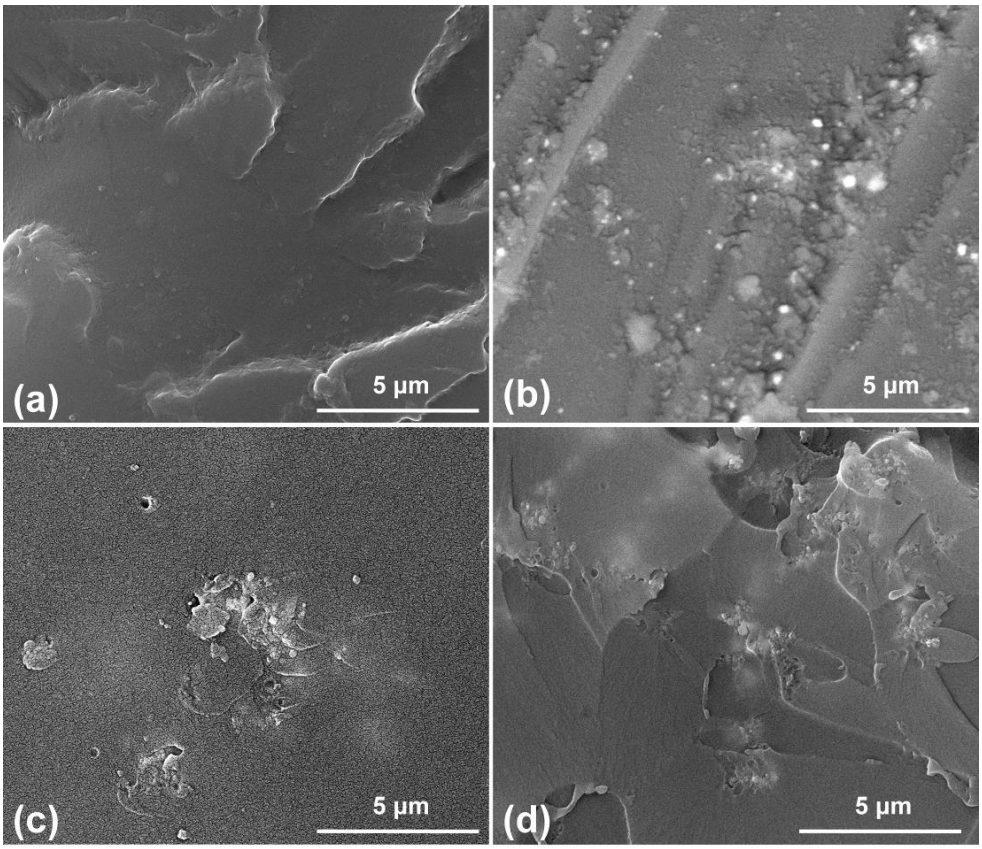

Fig. 3. SEM images of the fracture cross section of (a) Epoxy control sample, (b) Epoxy+0.5%nSiC, (c) Epoxy+1%nSiC, (d) Epoxy+2%nSiC

The cured epoxy resin control sample fracture cross-section presents both smooth areas and sharp edges and the morphology is characteristic for a polymer. The nanofilled sample fracture cross-section images illustrate that the nanoparticles are uniformly dispersed into the polymeric matrix, and they do not form large agglomeration areas.

The EDS analysis of one area from the 1% nSiC filled sample cross-section is shown in Fig. 4. The image illustrating Si element distribution map (Fig. 4-c) shows that Si is uniformly distributed on the whole visualized area, even in the areas where the SEM images does not suggest the presence of low dimension aggregates of nanoparticles.

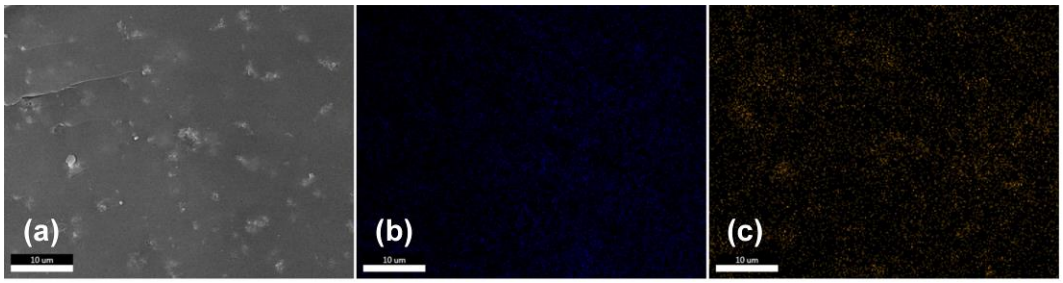

Fig. 4. EDS analysis of Epoxy+1%nSiC fracture area (a) cross-section image, (b) C elemental map, (c) Si elemental map

This suggests that the nSiC nanoparticles are strongly embedded into the epoxy matrix and uniformly distributed in the whole mass of the obtained composites.

3.3. Flexural testing

Flexural tests were performed on a minimum of 5 specimens per sample. As epoxy resin nature is brittle, most samples broke until standard deflection was reached. Table 1 presents the average results obtained after a mediation of the optimum specimens' results.

Table 1

Mechanical properties of the nanocomposites

Sample	Flexural strength (MPa)	Flexural modulus (GPa)	Extension (%)	Tensile strength (MPa)	Tensile modulus (GPa)	Extension (%)
Epoxy	99.66 ± 6.7	3.21 ± 0.6	3.74	46.27 ± 2.61	3.15 ± 0.21	2.58
Epoxy+0.5%nSiC	111.35 ± 7.49	3.21 ± 0.7	3.63	55.01 ± 0.4	3.18 ± 0.2	2.50
Epoxy+1%nSiC	125.32 ± 5.6	3.61 ± 0.04	3.52	59.19 ± 3.18	3.18 ± 0.12	2.39
Epoxy+2%nSiC	119 ± 9.12	3.4 ± 0.07	3.37	57.08 ± 0.45	3.2 ± 0.62	2,30

It can be noticed that both flexural strength and modulus of elasticity increase with the nanofiller addition compared to the unfilled control sample. As in the case of thermoset phenolic resin [26], it can be observed that the properties increase with nSiC content up to 1% by weight, while when adding 2% by weight, the values show a slight decrease compared to the previous nanofiller content, but remain higher than those of the unfilled sample. It can be noticed that flexural strength increments are more substantial compared to stiffness increments, the first are in the range of 12-26% compared to the control sample, while the latter are in the range of 0-12%.

3.4. Tensile testing

In the case of tensile testing, the average results of the obtained values are presented in table 1. The same trend as in the case of flexural test is observed.

Tensile strength increased by 20% when adding 0.5 wt.% nSiC, 28% when adding 1 wt.%, while 2 wt.% contents lead to 23% increase, slightly lower than the increments generated by lower contents of 1% by weight. Young's modulus of nanofilled samples shows a minor insignificant increase compared to the control sample.

As mentioned above, phenolic resin nanocomposites filled with 0.5, 1 and 2 wt.% nSiC presented the same trend in terms of mechanical properties. As both epoxy and phenolic resin are brittle materials with comparable viscosity, the explication of the obtained results has the same principle. As SEM images show, the SiC nanoparticles are uniformly embedded by the matrix, which creates an extended contact area between the two phases, allowing the nanometric phase to act as crack propagation reducer/stopper and leading to an increase of mechanical properties of the nanofilled matrix such as strength and stiffness.

3.5. Heat deflection temperature

Heat deflection temperature is a very important property for the materials application domain, as it is one of the parameters that define the temperature operating range.

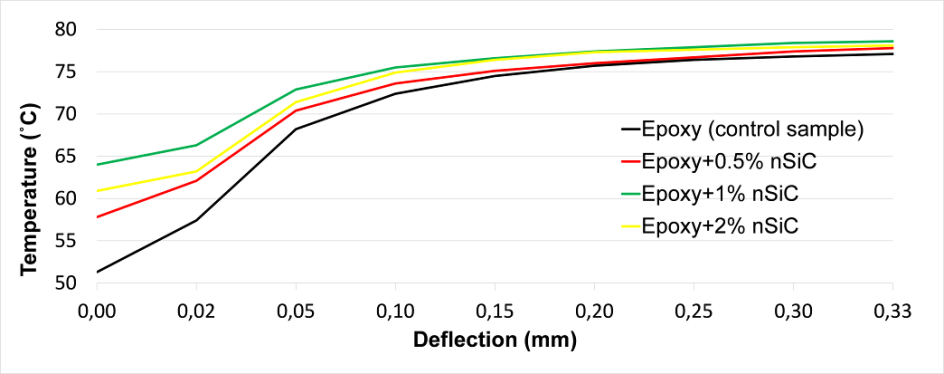

Fig. 5. Deflection- temperature curves during HDT test

The addition of nSiC to epoxy materials does not determine significant improvement of the final heat deflection temperature, but there is an important increase of the temperature corresponding to the starting point of the materials deflection, as Fig. 5 illustrates. The value of the initial temperature increases with the nSiC content increase in the matrix. The control sample starts to deflect at 51°C, while 2 wt.% nSiC filled samples start at 61°C, corresponding to a temperature-lag of approximately 20%.

3.6. Tribological testing

Tribological testing focused on the evaluation of the friction coefficient modification as a function of nSiC weight content into the epoxy matrix.

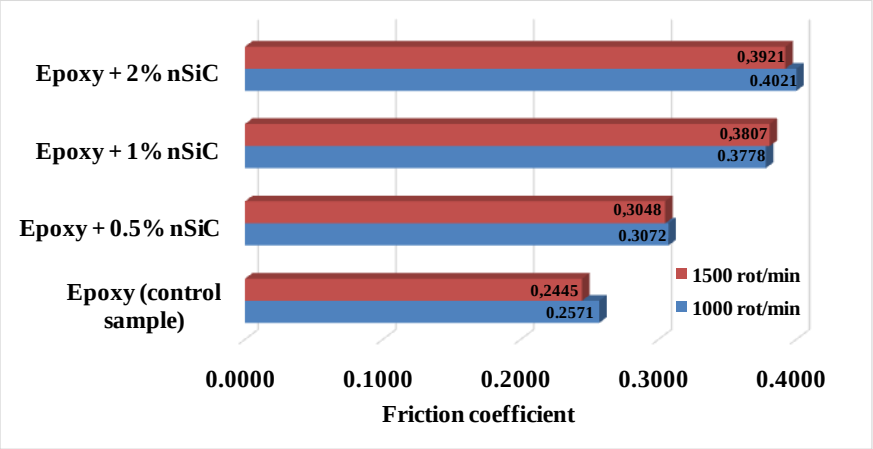

Fig. 6. Friction coefficient of nSiC filled epoxy measured at 1000 and 1500 rpm

The results clearly illustrate the abrasive nature of the nSiC nanofiller, the friction coefficient increases with nSiC content increase into the matrix, for both testing speed values. At 1000 rpm speed value, adding 0.5, 1 and 2 wt.% n SiC respectively generates an increase of friction coefficient by 20, 47 and 56% compared to the control sample, while at 1500 rpm, the same nSiC weight loadings generate increases by 25, 56 and 60% respectively. The results obtained when using 1 and 2% nSiC are very similar. However, taking into consideration SEM and EDS cross-section analysis and mechanical test results, it is clear that adding a higher nanofiller content (2 wt.%) increases the agglomeration tendency of nSiC and therefore, supports the occurrence of non-uniformity areas in the material. These non-uniformities can lead to different and inconsistent results, especially in terms of mechanical, but also tribological properties.

Overall, the results prove that adding nSiC can significantly improve the abrasive properties of epoxy resin, which extends this kind of material application domain towards higher friction materials.

4. Conclusions

The study presents the modification of mechanical, thermal and tribological properties of epoxy resin when adding nanometric silicon carbide in different weight contents. These preliminary results confirm the positive effect on nSiC presence, supporting the future research study focuses on developing advanced composite materials for aircraft and aerospace applications with improved polymeric matrix. Correlating mechanical and tribological test results with SEM analysis of the fracture cross-section, the results showed that 1 wt.% nSiC is the optimum content at which the nanofiller generates the highest mechanical and tribological properties improvement, with a minimum agglomeration tendency. The lower mechanical properties presented by 2 wt.%

nSiC based samples compared to the 1 wt.% samples suggest that at this content, the nanoparticles form more agglomeration areas that act as stress concentration sites that negatively affect strength and stiffness. The agglomeration phenomenon is probably not so intense in the 1 wt.% samples, the good embedding of the nanoparticles into the polymeric matrix, shown by SEM and EDS, allowing the nanofiller to act as crack propagation reducer agent. This allows the obtaining of more uniform structure materials with improved and consistent properties in mechanical, thermo-mechanical and tribological tests.

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R E F E R E N C E S

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