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Investigation on Wettability and Mechanical Properties of Novel Zinc Phosphate Glass-Based Epoxy Composites

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Abstract. Herein, we investigate the mechanical and wetting properties of composites-based epoxy matrix reinforced zinc phosphate glass particles at different volume fractions. The investigated composites were characterized using X-ray diffraction and Fourier-transform infrared spectroscopy. In addition, the morphology of composite was performed using scanning electron microscope tool. Moreover, the wettability of the composite performed by measuring water contact angle and surface energy. Contact angle findings revealed that the incorporation of zinc phosphate glass has significantly decreased with increasing filler concentration, while the surface energy values increased with filler concentration allowing improvement of the zinc phosphate glass-polymer interface. Besides, the investigated composites display enhanced mechanical properties for Young's modulus. In this study, the glass transition temperature has been determined using differential scanning calorimetry analysis which suggested that consolidated composites are significantly homogeneous and have good thermal stability.

1. Introduction

Epoxy resin is one of the most common thermosets, thanks to its high performance including excellent mechanical properties and reasonable chemical resistance in various fields. Nowadays, majority of wind turbine blades are composed of polymer matrix consolidated with carbon or glass [1]. The main advantages of these composites are low cost, lightweight and easy production. The rigidity and strength of polymers can be improved through the addition of stiffer and stronger spherical grains of glass powders in the polymer host matrix. Among all amorphous materials, phosphate glasses represent a class of optical glasses based (P_2O_5) and characterized by higher absorption coefficient, high thermal expansion coefficient, low glass transition temperature, low optical dispersion and good mechanical properties [2]. These outcomes promote zinc phosphate glass as a filler using polymer matrix and pave the way for composites with high performances in wide range of industrial applications.

This paper comprises new and advanced research on polymer-based composites where the focus is on the elaboration of bulk composite-based epoxy with different volume fractions of glass powders with the composition (45% mol. ZnO-55% mol. P_2O_5). Structural properties have been investigated by density and porosity measurements as well as XRD and IR-ATR techniques. These investigations were



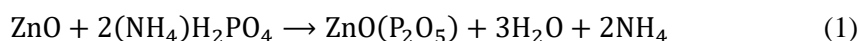
completed by morphological properties which were carried out through Scanning Electron Microscopy (SEM). Mechanical properties such as Young's modulus, tensile strength, elongation at break and elastic energy have been evaluated. Surface energies have been calculated via contact angles measurement and using Owens-Wendt approach. DSC analysis has been conducted to determine glass transition temperature for different volume fractions.

2. Experimental Details

2.1 Elaboration of zinc Phosphate Glass (ZP)

Powders of ammonium di-hydrogen phosphate ($\text{NH}_4\text{H}_2\text{PO}_4$ from Panreac type, 98%) and zinc oxide (ZnO from Panreac type, 99%) are mixed in appropriate molar proportion. The obtained powder was heated at 170 °C for one hour using alumina crucible in furnace, gradually increased to 300 °C and held for 12 hours for removing the traces of NH_3 and H_2O gases. The temperature was ultimately increased to 950 °C for getting supercooled liquid and kept for only half an hour to avoid any P_2O_5 volatilization. The melted powder was cooled in air at room temperature.

The glass formation was evidenced in the following equation:



45%mol. ZnO-55%mol. P_2O_5 composition was selected because it was tested as the most chemically stable glass composition.

2.2 Preparation of Zinc Phosphate Glass–Epoxy Composite

The used matrix is a bisphenol A epoxy F resin (Araldite) with a hardener HY956 (Ciba-Geigy). The measured density of epoxy at 22 °C is 1.14. The hardener was taken 25% and mixed with epoxy resin for getting homogeneous state. The fillers are the powder of phosphate glass (45%mol. ZnO-55%mol. P_2O_5). The powder of phosphate glass is mixed with prepared resin for obtaining homogeneous mixture. The formed viscous mixture was flowed in Teflon mold and mounted in a rotated system at 103 °C for 1 hour for avoiding sedimentation of the filler in oven.

The experimental density of neat epoxy and glass-epoxy composites was determined using the principle of Archimedes in accordance with ASTM D 792-91 through a Mettler Toledo (Columbus, OH), AJ 100 balance equipped with a density-determination kit. The samples were weighted before and after immersion in distilled water. Further investigation of structural characterization was done by performing X-ray diffraction. The diffractometer used for the assessment of bulk composites is of Bragg-Brentano geometry with copper anode of wavelength 1.5405 Å. Scans were performed over a 2-theta range between 10 and 70° with a step size of 0.02° and one second count time at each step. The prepared composites were also characterized by Fourier transform infrared attenuated total reflection (FT-IR/ATR) spectroscopy.

2.3 Tensile Strength Test

Stress-strain tests were performed using tensile strength testing machine.

2.4 Contact Angle and Surface Energy

The contact angle measurement was performed by capturing pictures through a camera. The angle is either directly measured by the software Screen Protractor, or calculated by software Analyzing Digital Pictures, by involving the dimensions of the drop as expressed in the following equation:

$$\theta = 2 * \text{Arctan} \left(\frac{2h}{d} \right) \quad (2)$$

where h and d are the height of drop and the length of drop respectively.

The surface energy of glass-epoxy composites was determined by the Owens–Wendt method using four solutions ethylene of glycol, glycerol, distilled water and ethanol [3].

2.5 Thermal Analysis of Zinc Phosphate Glass-Epoxy Composites

Thermal analysis of the investigated composites was carried out using differential scanning calorimetry (DSC), type SETARAM 121 under argon flow meter. The measurements were conducted at 20 °C/min using approximately 20mg of bulk composite in a Pt crucible.

3. Results and Discussion

3.1. Density and Porosity Results

Experimental and theoretical density curves are depicted in figure 1a. This figure shows that two curves (experimental and theoretical) increase steadily with the zinc phosphate glass inclusions and experimentally determined density is slightly less than calculated density. This phenomenon can be explained by the presence of certain porosity. Porosity rate of investigated composites is shown in figure 1b). It does not exceed 5% which means that elaborated composites are of good quality and homogeneous.

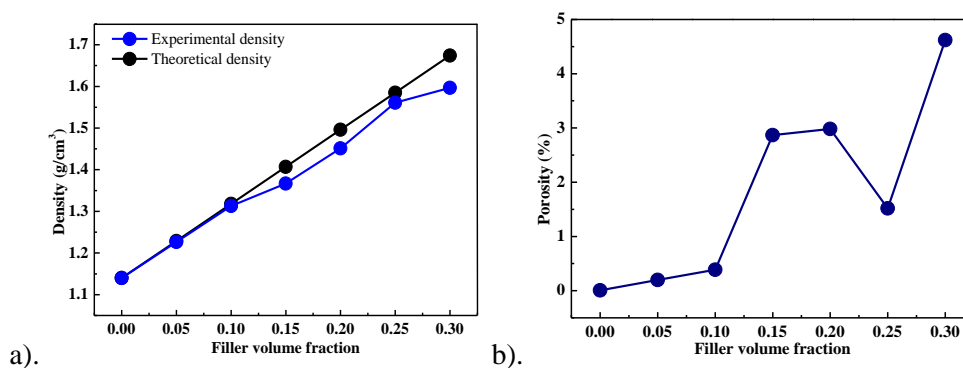


Figure 1. Variation of measured density and porosity of the Epoxy/ZP composite versus filler volume fraction. Solid lines represent calculated densities.

3.2. XRD Characterization

A set of X-ray diffraction spectra was performed on samples of zinc phosphate glass-epoxy composites to examine the effect of different incorporation levels of zinc phosphate glass on the structure of neat polymer matrix. The XRD patterns are gathered in Figure 2. Epoxy matrix shows a wide halo around $2\theta=18^\circ$ indicating amorphous nature of neat epoxy. The sequence of wide halo is still substantial in another collected XRD spectra with filler amount increasing which also suggests that the fillers are of amorphous state.

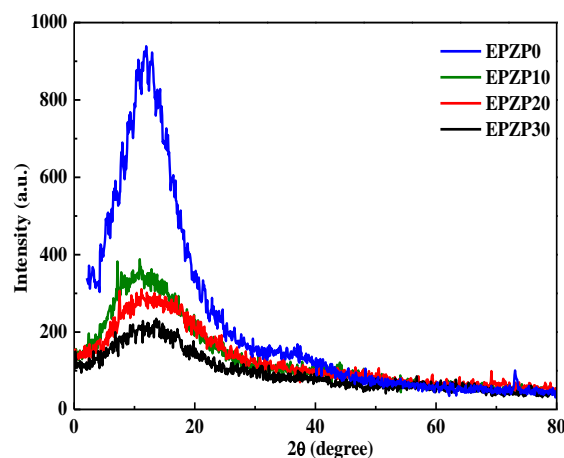


Figure 2. XRD results for the investigated composites.

3.3. Fourier Transform Infrared Spectroscopy Studies

FTIR spectroscopy was carried out for obtaining data of vibrational spectra that identified the role of zinc phosphate glass particles loaded in the epoxy matrix with different volume fraction. The infrared field ranging from 400 cm^{-1} to 4000 cm^{-1} correspond to molecules energy vibrations. The figure 3 compiled FTIR curves for EPZP samples loaded with different volume fraction of glass.

According to the attributions of detected bonds as represented beside the figure 3, the observed bonds correspond to the constituents of the neat epoxy matrix. No new band has been detected in the composites loaded with zinc phosphate glass as indicated in the figure 3. This suggests the non-existence of chemical bridges between the matrix and particles but only superimposed bonds indicating the dominant phases of the matrix are noticed.

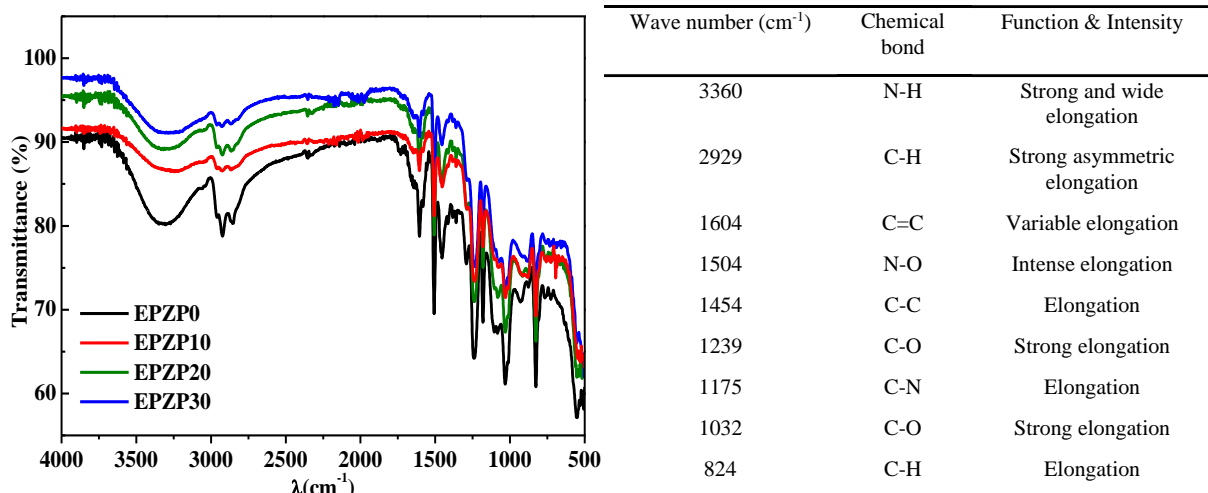
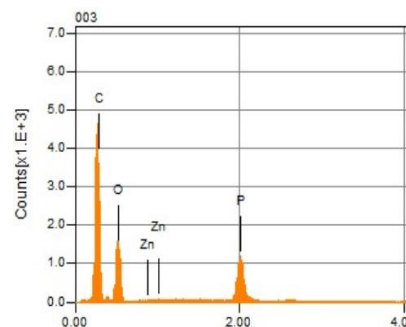
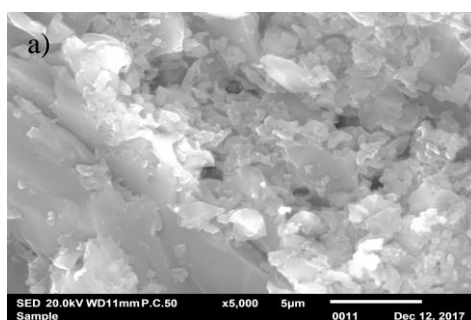


Figure 3. FTIR results of ZP/epoxy composites with different volume fractions and their attributions of FTIR bonds.

3.4. Morphology by SEM

SEM studies have been done to analyse the morphology and dispersion of zinc phosphate glass particles inside the polymeric matrix. The obtained images with EDX analysis using a magnification of (5000x) epoxy matrix loaded with 10 vol% and 30 vol% of zinc phosphate glass particles respectively (figure 4). It is clearly revealed from these pictures and EDX analysis that the composites are essential compounds with two components: matrix and incorporate fillers. The observed black–grey areas show two components epoxy matrix and zinc phosphate glass particles respectively. Moreover, these composites have a texture reflecting the presence of small void and pores inside the polymer matrix which is confirmed by measuring porosity of the elaborated composites.



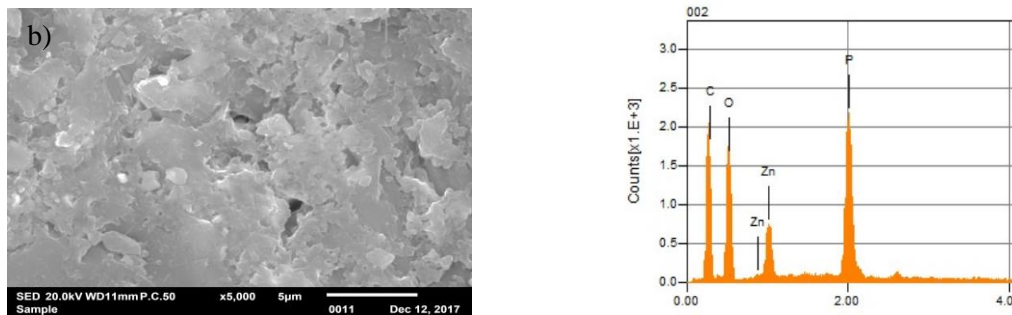


Figure 4. SEM image and EDX a) and b) respectively for the composite with 10% and 30% Vol of zinc phosphate glass particles respectively.

3.5. Mechanical Properties

Figure 5 shows the stress–strain curves at different strain rates for neat and loaded epoxy at 5, 15, 30 and 40% in volume percent of zinc phosphate glass powder subsequently. The stress–strain response is divided into two regions: elastic and plastic behaviors. The maximum stress point followed by abrupt failure at a strain of 6.5 % under quasi-static loading (2 mm/min) for neat epoxy system. However, it decreased in comparison with glass powder fillers to reach minimum value of 0.8% at 40% volume fraction (figure 6b). Figure 6a shows that Young’s modulus increases progressively with the increase of ZP filler volume fraction. The Young’s modulus has increased by magnitude of 161 % from 1157 MPa for the neat matrix to 3020.5 MPa at 40% vol.

Moreover, it is clear in figure 6a that higher loading level of ZP particles leads to noticeable decrease in the tensile strength of composites from 44 MPa for neat epoxy to 14.7 MPa at 40 Vol. % expressing loss of 66.6%. This phenomenon is more likely due to weak interfacial bonding between matrix and fillers [4] which has opened a new window for the investigation of glass-epoxy interaction knowing that ZP particles are hydrophilic whereas epoxy matrix is hydrophobic.

The damage occurred in the area of filler cluster and at filler/matrix interface can be generated in an earlier stage in the form of voids or cracks and results in the drop of tensile strength [5].

Strain at break is presented in figure 6b which exhibits significant decrease with the increase of ZP particles which is more probably due to decrease of ductility of the composite as compared to the neat epoxy. We conclude that the investigated composites become more brittle after using rigid particles than the neat matrix as demonstrated by the elastic energy calculation (figure 6b) which has expressed steady decrease with increasing ZP particles content.

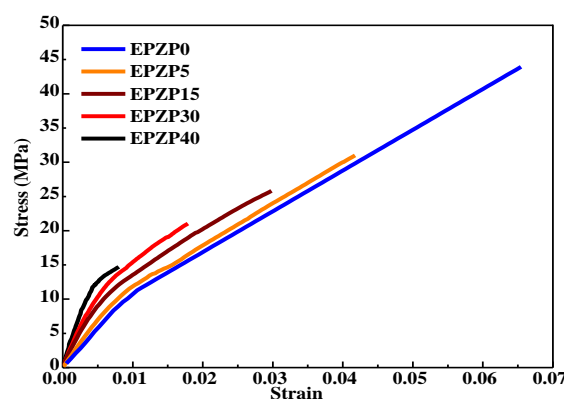
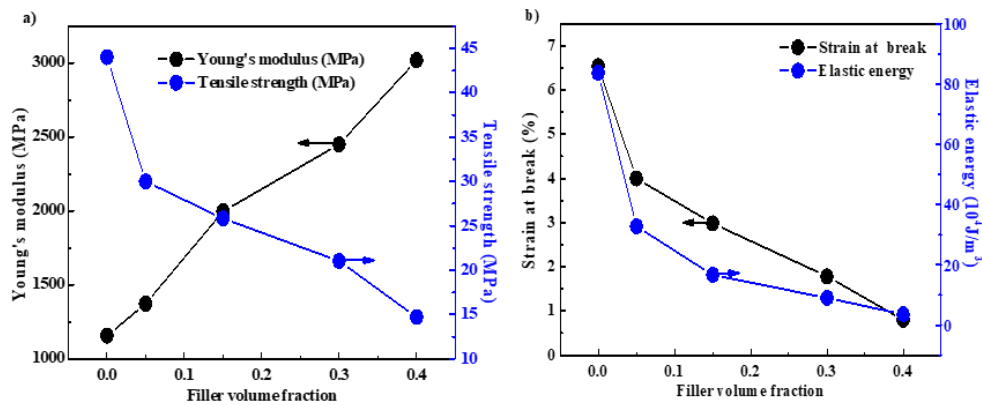
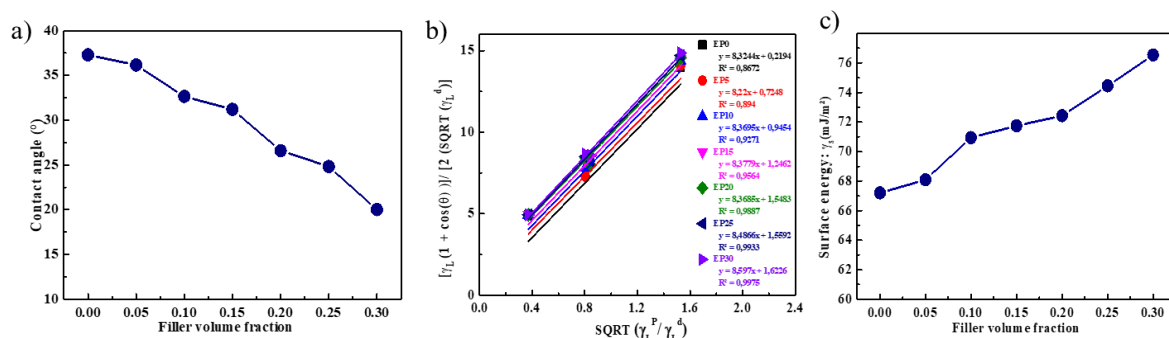


Figure 5. Stress-Strain curves of epoxy/ZP composites.**Figure 6.** The evolution of tensile properties of ZP/ Epoxy composites: a) b) Young's modulus & tensile strength; b) Strain at break & Elastic energy.

3.6. Wetting Properties

It was more convenient to calculate surface energy of the obtained composites for getting further information of zinc phosphate glass/epoxy interaction. Contact angle measurements were carried out knowing that contact angle is the perfect tool for the investigation of the composites surface energy. The measurement results of contact angle of different samples were performed by using distilled water are illustrated in figure 7a. As shown, the contact angle decreased with increasing of filler volume fraction which is coherent with expected results knowing that the ZP particles are hygroscopic. In order to calculate surface energy, the contact angle of composites was measured using following solvents: ethylene of glycol, glycerol, distilled water and ethanol. Furthermore, the regression of each sample was calculated using Owens-Wendt method as depicted in figure 7b. The surface energy was calculated for composite filled with different volume fraction of ZP particles as shown in figure 7c. The surface energy indicates a progressive increase, its initial value is 67.2 mJ/m^2 for the neat matrix and it reaches to 76.54 mJ/m^2 at higher loading level (30% vol.) representing 14% increase. Consequently, more ZP particles into the matrix maximizes the surface energy.

Indeed, the examination of surface energy reveals the wettability enhancement of the material which is due to the fact that ZP particles are hygroscopic. Therefore, the surface of composite becomes more and more wettable. When an organic polymer is mixed with an inorganic reinforcing filler, the polymer must wet the surfaces of the filler for promoting continuous phase boundaries. The strength across the phase boundaries or adhesion will naturally have effects on the stress-strain behavior of composites [6]. It was demonstrated previously while studying mechanical properties that Young's modulus E increases with the increase of filler. The wettability property has a significant effect on mechanical properties.

**Figure 7.** Graphical results of: a) Contact angle; b) Owens-Wendt regression; c) Surface energy.

3.7. Thermal Analysis DSC Method

The samples of neat epoxy along with investigated composites were examined by Differential Scanning Calorimetry technique (DSC). Figure 8 shows the obtained resulted curves of composites. All the curves indicate a smooth endothermic peak followed by an exothermic which corresponds to glass temperature. The latter seems to depend on the volume filler fraction of ZP particles. It is unusual that glass transition temperature is exhibiting significant increase with filler concentration of ZP particles. The interaction between filler-filler in polymer matrix is less at lower volume fraction filler when comparing at higher volume fraction filler. This has good relations with surface energy data. Indeed, lower loading leads to lower reinforcement effect of the fillers on the matrix chains [7] because ZP particles take place between the chains of epoxy matrix. Hence, the incorporated particles reduce the degrees of freedom of the chains and limit them for easier movement and pass to the amorphous phase under the temperature effect.

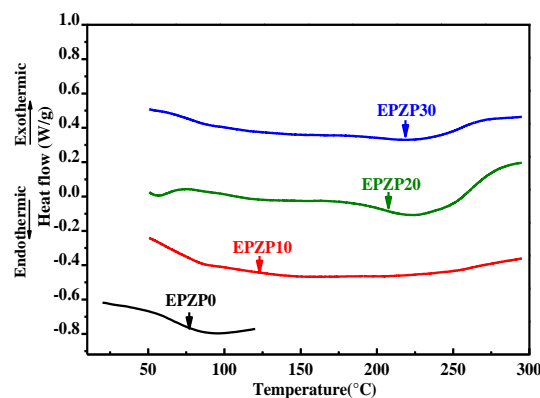


Figure 8. DSC results of ZP/epoxy composites.

4. Conclusion

This paper deals with structural, mechanical and thermal investigations of zinc phosphate glass-epoxy composites. Density and porosity calculations confirmed that composites are homogeneous. Furthermore, XRD and FTIR revealed that the addition of glass fillers within the matrix did not make any chemical change in epoxy matrix. Substantial bonds were seen based on the characteristics of the epoxy, hydroxyl and phenyl within the polymer. Additionally, the mechanical properties in terms of Young's modulus are enhanced by 161 % at 40vol% in comparison to the neat matrix. Therefore, strain at break and tensile strength showed decreasing behavior explained by that the ZP particles build stress concentration zones that cause interfacial debonding under stress. Besides, wetting properties of glass-epoxy interaction was examined via contact angle measurements which determined composites' surface energies. DSC analysis was performed for assessing the achieved results of structural study and surface energy investigations. These outcomes, as well as the lower costs and easy processing makes the investigated zinc phosphate glass cost effective, and great alternative material for the engineering of mechanical applications.

Acknowledgements

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