

Epoxy Matrix Composites Containing Urea Formaldehyde Waste Particulate Filler

Nurcan Calis Acikbas¹  · Gokhan Acikbas²

Received: 18 April 2016 / Accepted: 29 July 2016 / Published online: 6 August 2016
© Springer Science+Business Media Dordrecht 2016

Abstract In the present study, industrial toilet seat wastes which contain 70 wt% urea formaldehyde and 30 wt% cellulose were used as a particulate reinforcement in epoxy matrix and their mechanical and physical properties were investigated. The usage of urea formaldehyde and cellulose mixture as filler of polymer composite materials is a novel study. Initially toilet seat wastes were ground and particles in the required size range were obtained. The characterization of waste particulate filler was carried out by scanning electron microscopy, X-ray diffraction techniques and He gas pycnometer. Optimum concentration of the filler was determined as a maximum of 40 wt% because of the increase in viscosity at higher waste concentration. The effect of particle size on mechanical properties such as hardness, bending strength and bending modulus was evaluated. These are the key properties most likely to be affected by incorporation of reinforcement phase. Experimental results showed that bending modulus and hardness increased and bending strength decreased with filler addition into epoxy resin. With the increase in filler content both porosity and bending strength decreased and bending modulus increased. On the other hand increase in particle size led to the enhancement of bending strength and bending modulus while accompanied with a decrease in porosity. As a result, incorporation of urea formaldehyde wastes basically resulted in the reinforcement of the epoxy matrix. This allows for the recycling of hardly recoverable

thermosetting residues as well as improving some mechanical properties of the composites.

Keywords Epoxy · Urea formaldehyde waste · Cellulose · Bending strength · Hardness

Introduction

Basically, a composite material is a material system composed of two or more macro constituents that differ in shape and chemical composition and which are insoluble in each other. It is possible to achieve combinations of properties with composites which are in general not attainable with metals, ceramics, or polymers alone. Composites contain two or more different phases which are matrix and reinforcement. The continuous phase that is present in the greater quantity in the composite is termed the matrix, which may be ceramic, metal or polymer. Matrix may be ceramic, metal or polymer. The second constituent is called as reinforcement, since it enhances mechanical properties of the matrix. The geometry of the reinforcement is one of the major parameters in determining the effectiveness of the reinforcement. In other words, the shape or dimensions of reinforcement determine the mechanical properties. Reinforcements may have particle, short or long fiber form. Another important parameter that determines mechanical properties is interface bonding. The load acting on the matrix has to be transferred to the reinforcement via the interface. It is possible to produce controllable properties by changing an appropriate amount of constituents [1–5].

Many common examples of composite materials can be found in the world such as wood and bone as natural composites. On the other side modern composite materials

✉ Nurcan Calis Acikbas
nurcan.acikbas@bilecik.edu.tr

¹ Department of Metallurgical and Materials Engineering, Bilecik S.E. University, Bilecik, Turkey

² Metallurgy Program, Vocational School, Bilecik S.E. University, Bilecik, Turkey

are used in aerospace, transport, construction industry, shipbuilding industry, anti-corrosion equipment, electrical and electronic industry, all kinds of sports equipment, agriculture and fisheries and mechanical manufacturing [5–7].

Polymer matrix composites are the most widely used among the composites [8, 9]. For decades different kinds of waste materials have been successfully used as filler in polymer matrix composites. This not only reduces the production costs but also reduces environmental pollution by utilizing waste materials. A huge amount of waste is released by industries, which leads to serious environmental problems. Eco-consciousness and government regulations compelled the researchers to study the use of industrial wastes as a reinforcement phase in polymer matrix composites. Industrial wastes as a reinforcement phase provide low cost, ease of manufacturing and high mechanical and other properties [10]. Industrial waste such as metallurgical slag, foundry slag, waste glass, waste rubber, gypsum-fiber waste, red mud, blast furnace slag, fly ash, flue dust, and carbon black waste e.g. has been used as filler to reinforce polymer [11–25].

A number of studies about the use of waste as filler in epoxy matrix composites are available in the literature. In our previous study, ceramic wastes which were porcelain and boron were used in epoxy matrix in order to reduce resin cost and improve mechanical properties. The results showed that addition of 60 wt% of ceramic waste was optimum content for casting. Epoxy matrix composites with porcelain waste showed better properties than boron waste added composites due to more porosity evolution [26]. Biswas and coworkers studied industrial wastes that used red mud and copper slag particles as filler material with bamboo fiber and glass fiber reinforced epoxy matrix composites. The results showed that the density increased with the red mud and copper slag filling for both bamboo and glass fiber reinforced composites. With the addition of 10 % red mud, the elastic modulus increased. The addition of filler did not affect the strength since the adhesion between the fibers, filler and matrix was poor, and the stress could not be transferred from the matrix to the fibers. Flexural strength increased with the addition of red mud with the maximum value of 164 MPa with a filler content of 20 wt%. A gradual decrease in the strength was found for bamboo and glass fiber reinforced composites with the increase in the copper slag content. Tensile properties were getting inferior with the incorporation of copper slag particles in the epoxy-glass composites, but tensile modulus improved for bamboo reinforced epoxy resin. The flexural strength decreased with increase in the copper slag content for bamboo epoxy composites. Impact strength increased linearly with copper slag content increasing from 0 to 20 wt% [27]. Barbuta and coworkers studied polymer concrete by adding different types of wastes, such as

argillaceous powder, calcareous powder, marble powder and fly ash in epoxy matrix. It was found that the calcareous and fly ash addition in polymer concrete mix improved the mechanical properties [28]. Fombuena et al. [29] studied the addition of calcium carbonate from seashells in epoxy resin. The results showed that incorporation of 30 wt% of seashell increased mechanical properties as flexural modulus (over 50 %) and hardness Shore-D (over 6 %) and thermal properties as an increase around 13 % in glass transition temperature. It was concluded that this is an effective method to increase mechanical properties of bio-composite and to reduce the residue of seashells from industrial production. Goh et al. [30] used fly ash waste as filler in epoxy resin to enhance the mechanical strength of the composite. Surface modification was performed on fly ash to improve the interfacial bond of the filler and epoxy resin. The results revealed that unmodified fly ash weakened both flexural and tensile strengths. On the other hand, addition of mesoporous silica treated fly ash into epoxy matrix resulted in improved mechanical properties with respect to unmodified and silanized fly ash, with remarkable enhancements of flexural and tensile strengths and modulus. Purohit and Satapathy studied wear of epoxy matrix composites containing LD sludge with different content (0, 5, 10, 15 and 20 wt%). The results showed that sludge content and sliding velocity affected the specific wear rate more significantly than normal load and sliding distance. Also, with increase in sludge content the wear rate of the composite decreases for a constant sliding velocity [31]. Prithvirajan and coworkers studied agricultural residues such as rice husk and coir pith particulates with varying amount (10–50 wt%) as reinforcement in epoxy matrix as an alternative to wood and plastic based composites. They stated that the addition of rice husk and coir pith particulates increased the mechanical properties of the composite [32]. Martins et al. [33] developed epoxy composites incorporated with 30 % in volume of continuous sugarcane bagasse fibers and evaluated their mechanical properties. The results revealed that bagasse fibers improved the elastic modulus but a decrease strength and ductility.

Despite the fact that considerable research is being carried out on waste containing epoxy matrix, no study on reuse of thermosetting polymers like urea–formaldehyde (UF) as a waste and filler in epoxy is available. Recycling of thermoset polymer is more difficult since they have a cross-linked structure and cannot be remolded in contrast to thermoplastics which can be easily remelted [34]. Toilet seats are produced basically by using a molding composition which is a mixture of UF and cellulose in the form of granules. The mixture was cured under heat and pressure with the compression molding method [35]. Generally, the amount of defective products is in between 3 and 10 % during production. The 3–10 % defective production of a typical company corresponds to a waste that amounts to

10–30 tons per year. Perceived lack of recyclability is now increasingly important and seen as the key barrier to the development or even continued use of thermoset composites in some markets. Therefore recycling of thermoset polymers is an important issue in the world [36].

Toilet seats are an industrial waste which contains 70 wt% urea formaldehyde and 30 wt% cellulose. The major advantages of the waste as a reinforcement phase are low density, fiber like microstructure of cellulose. Recently, Barari and co-workers has used the plant derived cellulose nano fibers as reinforcement in epoxy matrix [37]. The results showed that addition of cellulose fiber provided enhanced mechanical and tribological properties compared to pure epoxy samples. Addition of cellulose may increase the tribological performance of toilet seat waste added to epoxy composite and hence this kind of composites has a potential to be used as engineering parts which are exposed to wear and friction. However, cellulose has a hydrophilic character and hence adherence between the matrix and cellulose is poor. In order to improve adherence to polymeric resin functionalization of cellulose by direct polymerization of monomers groups is one of the most useful processes [38].

Formaldehyde-based materials are key to the manufacture of automobiles, and used to make components for the transmission, electrical system, engine block, door panels, axles and brake shoes. The value of sales of formaldehyde and derivative products was over \$145 billion in 2003, about 1.2 % of the gross domestic product (GDP) of the United States and Canada. Approximately 1 million metric tons of urea–formaldehyde are produced every year. Over 70 % of this production is then put into use by the forest industry products for bonding particle board (61 %), medium density fiberboard (27 %), hardwood plywood (5 %), and laminating adhesive (7 %) [39].

To the best of our knowledge there is no study available for use of toilet seats as the reinforcement phase. Therefore in this study, the use of hardly recoverable thermosetting toilet seat waste which contains UF and cellulose as a reinforcement phase was evaluated in epoxy resin. The effect of filler content and particle size on mechanical and physical properties was determined. Such understanding is necessary, due to growing interest and new potential applications. The study was also aimed at eliminating the waste from the environment.

Experimental Studies

Materials and Methodology

The matrix material consists of epoxy resin and hardener was supplied by Smoth-on Limited, Canada. The mixing ratio of the epoxy and hardener was taken as 73:27 by

weight. Toilet seats are an industrial waste, taken from ceramic company, Bilecik, Turkey. Waste material was ground with a Fritsch Vibrating Cup Mill Pulverisette 9 for 2 min at 900 rpm. After that ground waste was dry sieved for 5 min with Fritsch Vibratory Sieve Shaker Analysette 3 Spartan to obtain a particle size in the range of 45, 90 and 150 μm . The schematic presentation of preparation of waste and characterization is given in Fig. 1. The theoretical density of waste was measured by Micromeritics Accupyc II 1340 model He-gas pycnometer and found as a 1.47 g/cm^3 . The types of phases were determined by means of X-ray diffraction analyses (XRD-Panalytical, Empryan with Cu-K α radiation). Figure 2 represents the XRD pattern of waste material. The graph showed that guanidine (1, 3, 4 and 5 coded arrows indicated the phases) and dinitrogen tetroxide (2 and 4 coded arrows indicated the phases) phases were available in waste material. Secondary electron scanning electron microscopy (Zeiss Supra 40 VP FEG-SEM) images of waste material are given in Fig. 3. Particles are of irregular shape and of different size after grinding. The particle shape affects the process ability of composite and some mechanical properties.

Monolithic epoxy resin and composite materials with different epoxy: waste ratio (wt%. 70:30, 60:40, 50:50) and different waste: particle size was prepared (Table 1). The coding is as follows: the first two figures following W represent the particle size and the last two figures represent the waste ratio. The process flow chart was given in Fig. 4. Polymer matrix composites were produced with the casting method. Before casting, the mold is coated with lubricant that is polyvinyl alcohol (PVA) to easily remove the composites from the mold. Epoxy resin, hardener and waste particulates were blended together at 500, 1000 and 1500 rpm for 5 min, respectively. The purpose of the slow mixing rate in the beginning is to avoid the loss of filler powder by dusting. Once the filler is incorporated into the liquid polymer the mixing rate could be increased stepwise to obtain homogeneous mixing and avoiding the vortex reaching the propeller. After that the blended mixture was subjected to vacuum application in a desiccator to remove the bubbles and prevent subsequent pores in the composite. The blended mixtures were poured into silicon molds. The epoxy resin-waste particulate blend was left for 24 h in the mold at room temperature for curing after which the epoxy resin-waste particulate composite was carefully removed from the mold.

In order to determine the theoretical density of the composite, the theoretical densities of the waste and epoxy resin were used. Theoretical density of composite was calculated by using law of mixture as following equation [1–6]. In order to calculate the theoretical density of the composite, weight fractions were converted to volume fractions.

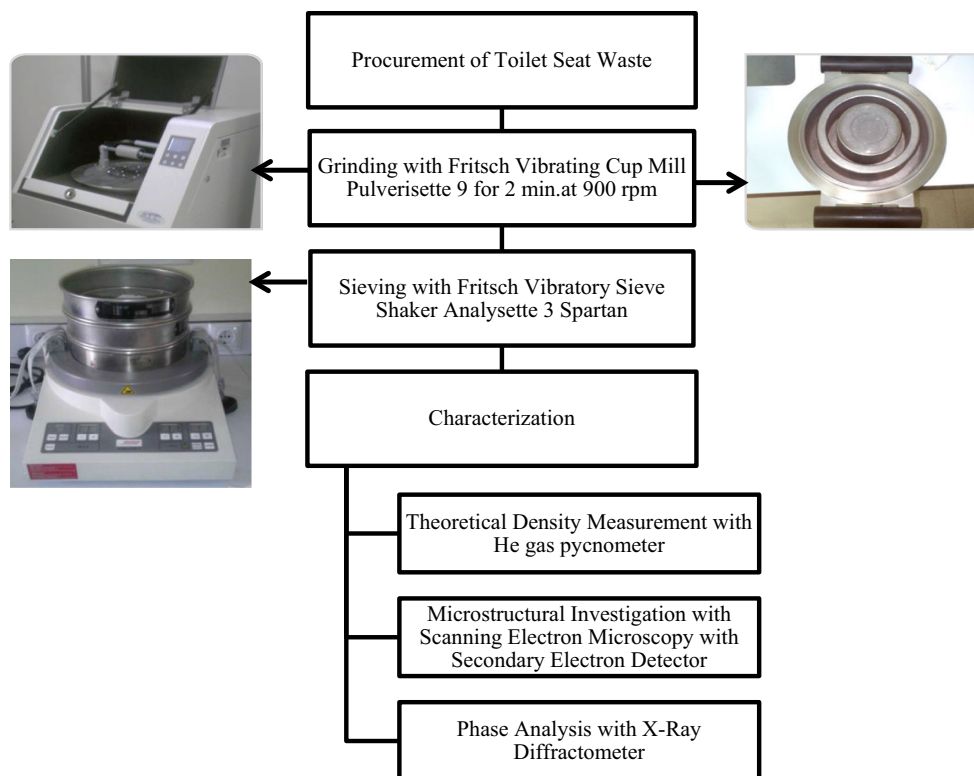


Fig. 1 Schematic presentation of waste preparation and characterization

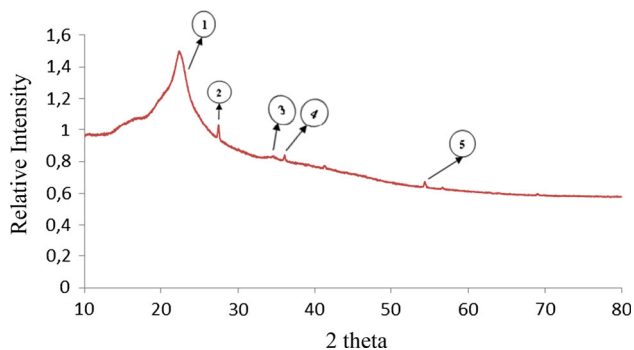


Fig. 2 XRD pattern of waste material (1, 3, 4 and 5 peaks: Guanidine $\text{HNC}(\text{NH}_2)_2$, 2 and 4 peaks: Dinitrogen tetraoxide N_2O_4)

$$\rho_c = \rho_m \cdot V_m + \rho_r \cdot V_r \quad (1)$$

where ρ_c , theoretical density of composite, ρ_m , theoretical density of matrix, ρ_r , theoretical density of reinforcement, V_m , volume fraction of matrix, V_r , volume fraction of reinforcement.

The microstructural investigation of the fractured surfaces of samples was performed by means of a scanning electron microscope (SEM-Zeiss Supra 40VP). The hardness of plastics is commonly measured by a shoremeter. The method measures the resistance of plastics toward indentation. The Shore-D hardness was measured of samples with 5×5 cm in size. At least five measurements

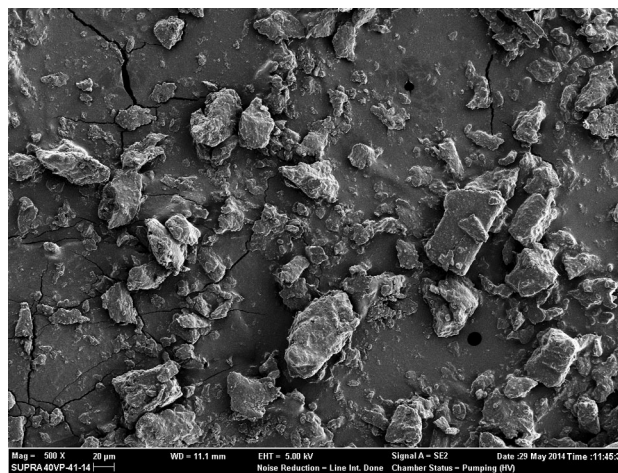


Fig. 3 SE-SEM image of waste material

were made for each sample and average was taken. 3 point-bending strength tests of samples was done according to TS 985 EN ISO 178 standard. At least three samples were used for a test. After that bending strength, elongation, bending modulus were determined. The bending modulus of composites was calculated using the following equation.

$$E = \frac{L^3}{4WD^3} \times m \quad (2)$$

Table 1 Specifications of prepared composite materials

Code	Epoxy: waste ratio (wt%)	Particle size of waste (µm)
W4530	70:30	<45
W4540	60:40	<45
W4550	50:50	<45
W9030	70:30	<90
W9040	60:40	<90
W15040	60:40	<150

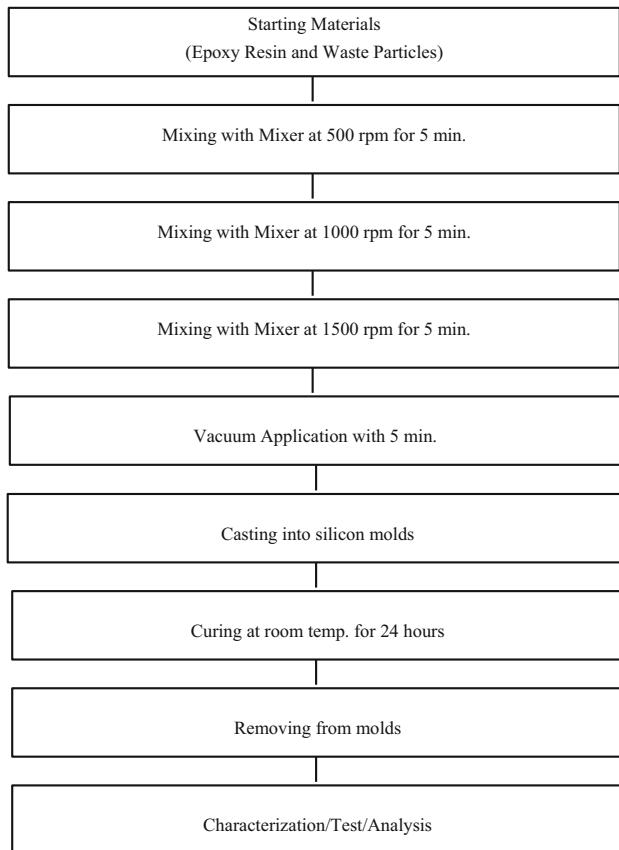


Fig. 4 Composite production process flow chart

where E is the bending modulus, m is slope, L is the distance between the span, W is the width and D is thickness of the tested sample.

The Archimedes principle was used to measure the density and porosity of the samples. Bulk density, %open porosity, %theoretical density and %total porosity were calculated using the following equations.

$$\text{Bulk Density} = \frac{W_1}{W_3 - W_2} \times \rho_{\text{water}} \tag{3}$$

$$\% \text{Open Porosity} = \frac{W_3 - W_1}{W_3 - W_2} \times 100$$

$$\%T.D. = \frac{B.D}{T.D} \times 100$$

$$\% \text{Total Porosity} = 100 - \%T.D.$$

where, W_1 is dry weight, W_2 is wet weight suspended in water, W_3 is wet weight, B.D. is Bulk Density, T.D. is Theoretical Density of samples.

Results

Determination of Optimum Epoxy:Waste Ratio

One of the objectives of the study is producing low cost composites. Therefore the waste amount should be as high as possible. For the determination of optimum epoxy:waste ratio for casting, the particle size of waste was kept constant as 45 µm. Varying amounts of waste (epoxy:waste ratio 70:30, 60:40, 50:50) were added into epoxy resin in order to observe the processing ability of the mixture (epoxy and waste). Some observations about the casting behavior of composite mixture were given in Table 2. With the addition of 30 % waste, easy mixing, little porosity evolution, no lamination problem, low viscosity and easy casting were obtained. With an increase in waste amount from 30 to 40 wt%, the blend was still mixable, the viscosity was low, there was little evolution of porosity, no lamination, low viscosity and the mixture was castable. However, an increase in waste content led to remarkable increase in viscosity. With a further increase in the waste amount to 50 wt%, the mixing was getting worse, there was high porosity evolution due to the difficulty of processing the mixture, no lamination problems, but very high viscosity and casting was difficult. Due to significant loss of fluidity, additions over 40 wt% have not been efficient. Therefore it was found that a 60 epoxy: 40 waste ratio was optimum to get low cost composites with processing ability. A longer time of mixing or a sonication process may be tried to provide proper distribution of the waste in the polymer matrix [40].

Physical Properties of Composites

Table 3 shows the bulk density, theoretical density, % theoretical density and vol %total porosity of composite materials. The bulk densities of composite are changing between the 1.17 for the W4530 to 1.25 g/cm³ for the W15040. Bulk density consists of open and closed pores. So if the material has high bulk density, total pore amount will be low. Since the W15040 coded composite has bigger waste particle size (150 µm) its process ability is better and hence has the lowest porosity evolution and highest bulk density value. With an increase in waste content and

Table 2 Observations about casting behaviour of epoxy:waste mixtures

Sample	W4530	W4540	W4550
Observations	Pore formation is little No lamination Low viscosity Easy casting Homogeneous distribution of particles	Pore formation is available No lamination Medium viscosity Homogeneous distribution of particles	High pore formation No lamination High viscosity Difficult casting Homogeneous distribution of particles

Table 3 Bulk density, theoretical density, % theoretical density and % total porosity of composite materials

	W4530	W4540	W4550	W9030	W9040	W15040
Bulk density (g/cm ³)	1.17	1.18	1.19	1.19	1.22	1.25
Theoretical density	1.48	1.49	1.49	1.48	1.49	1.49
% T.D.	77.34	78.99	79.40	80.65	81.61	83.42
% Total porosity	22.66	21.01	20.60	19.35	18.39	16.58

particle size of added waste material, the bulk density, theoretical density, % theoretical density values increased and total porosity level decreased. If the particle size is less, the particles have higher surface area and process ability is worse. Therefore a composite consisting of fine particle size of waste causes more porosity evolution and less %theoretical density. Representative SEM images of W4540 and W15040 coded composites were given in Fig. 5. As seen from the SEM images, the fine particle containing composite (W4540) has a higher porosity than coarse particle containing composite (W15040). The results corresponded with the total porosity values which were obtained from the Archimedes principle.

Shore-D Hardness

The Shore-D hardness results of samples are given in Table 4. The Shore hardness of the waste material was

measured as 94 ± 0.3 . Addition of waste particles into epoxy resin caused an increase in hardness, so monolithic epoxy material showed lower hardness than composite material. The hardness value remained almost constant when there was an increase in the amount of waste particles in the composite (Fig. 6). On the other hand the hardness value showed only a slight increase when larger particles (<150 micron) are used instead of fine particles (<45 μm) (Fig. 7).

Porosity affects the hardness of the composite. In order to relate the shore hardness with porosity, microstructural investigation of composites was carried out and compared with the total porosity values which were obtained from the Archimedes principle applied to the composite. As seen from the SEM images (Fig. 7), fine particle containing composite (W4540) had a higher porosity (21.01 %) than coarse particle containing composite (16.58 % for W15040) since they have higher surface area and were

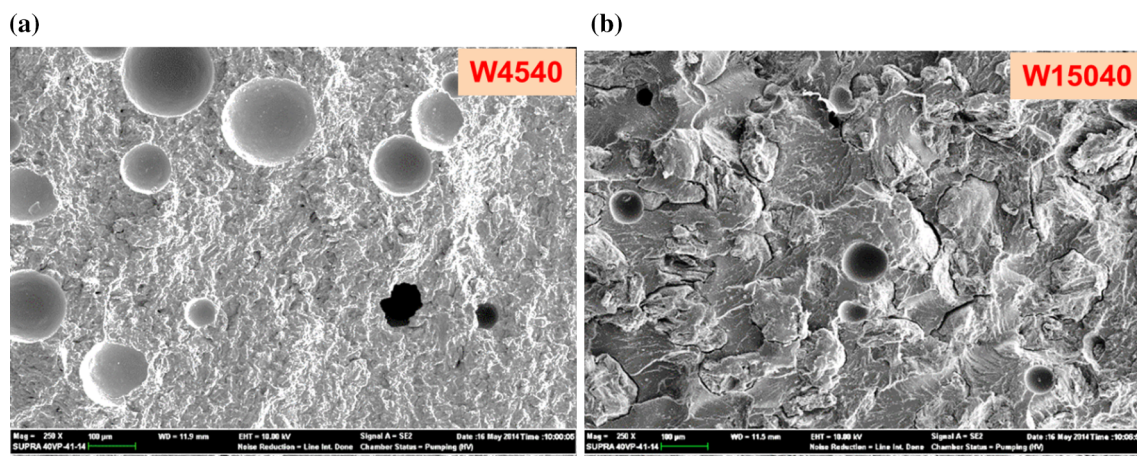
**Fig. 5** Representative SEM-SE images of composites **a** W4540, **b** W15040

Table 4 Shore-D hardness values of composites

Sample code	Shore hardness
Epoxy	80.50 ± 4.9
W4530	85.70 ± 0.3
W4540	85.40 ± 0.1
W4550	85.80 ± 0.4
W9030	86.60 ± 1.2
W9040	86.90 ± 1.5
W15040	87.00 ± 1.6

Table 5 Bending strength and bending modulus values of composites

	Bending strength (MPa)	Bending modulus (GPa)
Epoxy	106.00	2.13
W4530	62.94	3.46
W4540	65.25	3.74
W4550	66.01	4.57
W9030	69.90	3.31
W9040	73.11	4.33
W15040	91.52	4.57

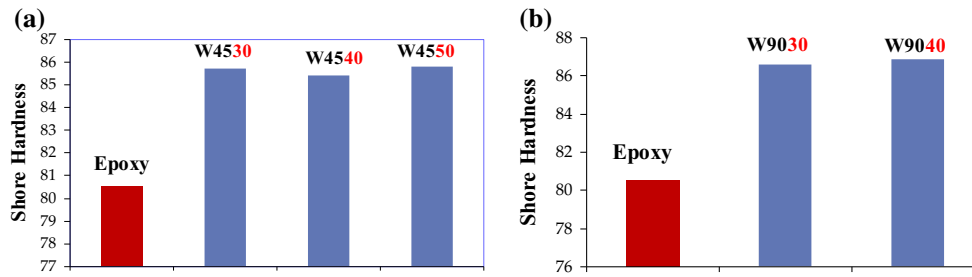
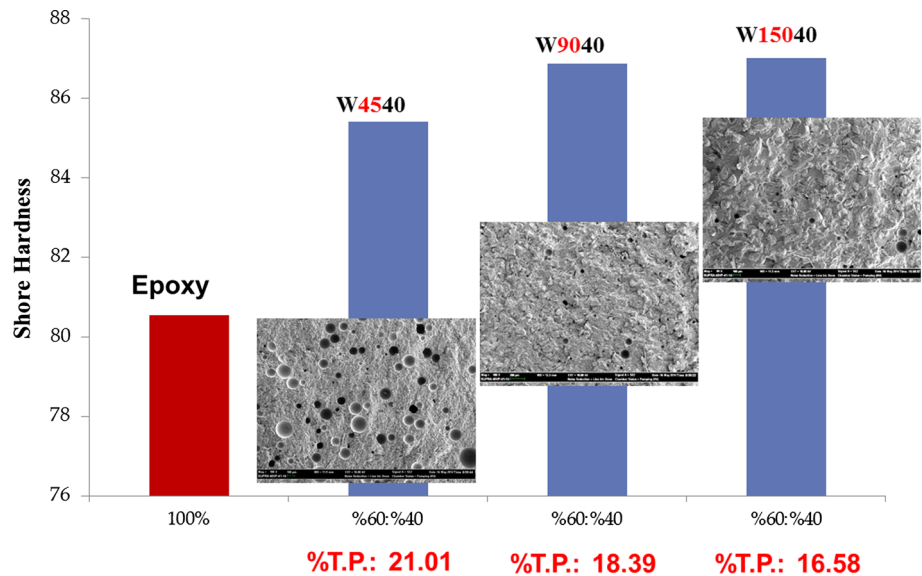


Fig. 6 Hardness-waste content relationship for **a** W45 coded composites, **b** W90 coded composites

Fig. 7 Hardness-waste particle size relationship (%T.P. is total porosity percentages)



difficult to process. As a result, increase in porosity led to decrease in hardness. Similar results were obtained in the literature about the effect of porosity on hardness [41].

Bending Strength and Bending Modulus

Table 5 shows the bending strength and bending modulus of composite samples. Bending strength values are changing between 62.94 MPa for W4550 and 106 MPa for the epoxy matrix. Addition of waste particles into epoxy resin caused a decrease in bending strength. On the other hand with the increase in waste amount, the bending

strength was increased slightly. As seen from the graphs (Fig. 8), addition of hard particles into epoxy resin led to a decrease in bending strength due to waste particles providing more brittleness. When the waste particle size was fine the bending strength remained relatively constant with the increasing amount of the waste content (Fig. 8). Bending strength values changed between 62.94 to 66.01 MPa for W45 coded samples. This might result from the homogeneous distribution of waste particles in the epoxy resin. Also the interface bond strength between the matrix and reinforcement might be similar with different waste amounts.

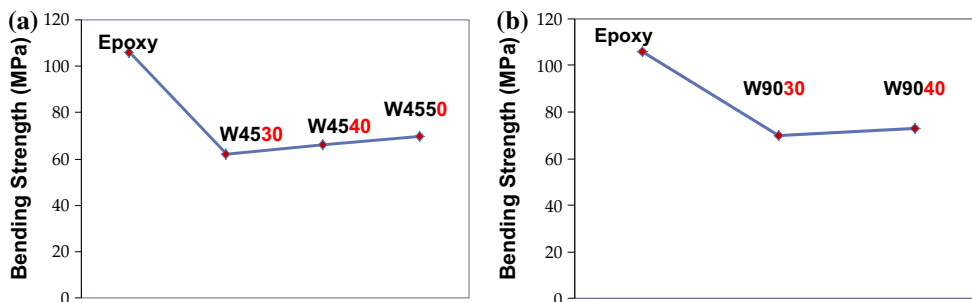


Fig. 8 Bending strength-waste ratio relationship for a W40 coded composites, b W90 coded composites

The use of coarser waste particle size led to the improvement of bending strength (Fig. 9). While W4540 sample had 65.25 MPa bending strength value, W9040 and W15040 coded composites had 73.11 and 91.52 MPa bending strength values, respectively. With use of coarse particles instead of fine particles the bending strength showed an increase about 12 and 25 %, respectively. This may be affected by the porosity content and the nature of

the reinforcement phase. An increase in filler particle size leads to a decrease in filler volume in the composite. Therefore the material shows similar characteristics with epoxy matrix which have higher bending strength than reinforced composite.

An increase in porosity in the microstructure results in lowering of bending strength. SEM images of W4540, W9040 and W15040 sample’s fracture surfaces are given

Fig. 9 Bending strength-waste particle size relationship

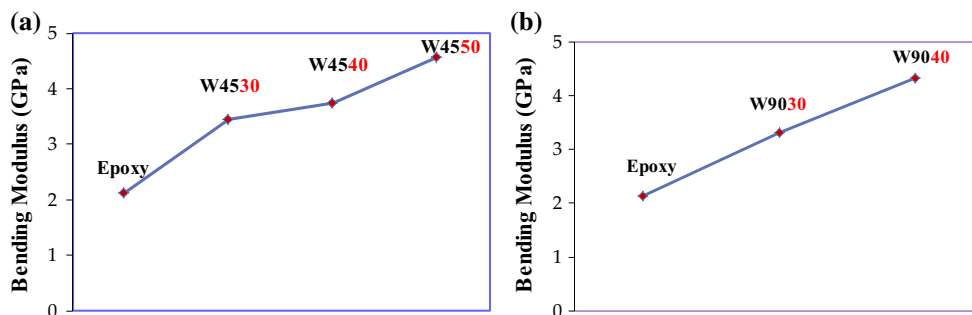
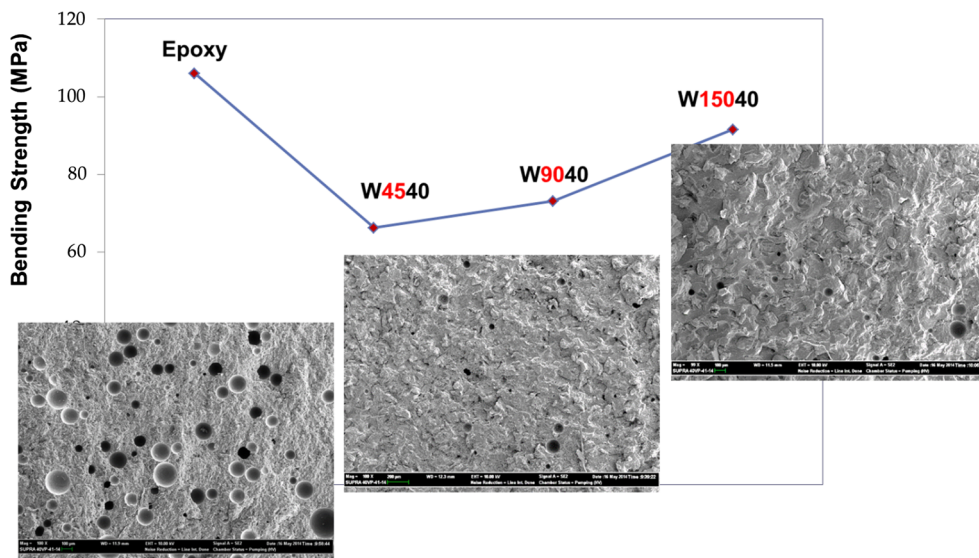


Fig. 10 Bending modulus-waste content relationship a W40 coded composites, b W90 coded composites

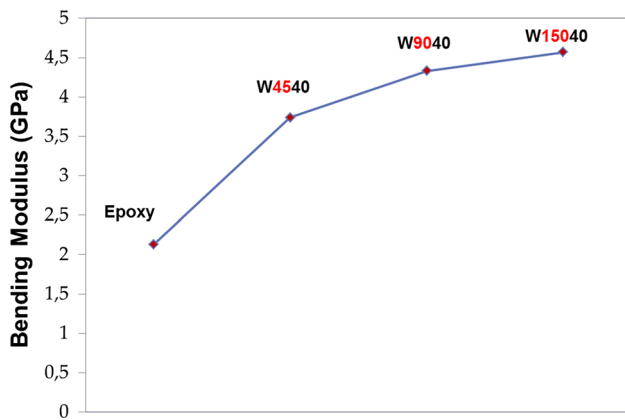


Fig. 11 Bending modulus, waste particle size relationship

in Fig. 9. From the images it was obvious that porosity level was getting low with the increase in particle size of the waste. On the other hand, cellulose with fiber like structure in the waste material may result in enhancement of bending strength. The highest bending strength was obtained from W15040 composite.

Bending modulus enhanced with the addition of hard waste particles into epoxy resin. Besides an increase in waste ratio the bending modulus showed an increased trend (Fig. 10). The particle addition led to an increase in rigidity revealed by an enhancing bending modulus observation. Similarly, porosity also affected the bending modulus. A decrease in porosity in the microstructure caused an increase in bending modulus. A similar trend was also observed for W90 compositions. Use of coarse particle size of waste gives higher bending modulus than finer ones. It can be related to again porosity amount and hence process ability of epoxy-waste mixture (Fig. 11).

Conclusions

The optimum pourable epoxy:waste ratio was determined as 60:40 wt%. The results revealed that bending modulus and hardness increased and bending strength decreased with filler addition into epoxy resin. When the filler content increased, both the porosity and the bending strength decreased, while the bending modulus increased. On the other hand increase in particle size led to the enhancement of bending strength and bending modulus accompanied with a decrease in porosity. The incorporation of urea formaldehyde wastes basically resulted in the reinforcement of the epoxy matrix. This allows for the recycling of hardly recoverable thermosetting residues as well as improving some mechanical properties of the composites.

References

1. Matthews, F.L., Rawlings, R.D.: Composite Materials Engineering and Science. Woodhead Publishing, Cambridge (1994)
2. Hull, D.: An Introduction to Composite Materials, Cambridge Solid State Science Series. Cambridge University Press, Cambridge (1981)
3. Chawla, K.K.: Composite Materials: Science and Engineering. Springer, Berlin (1998)
4. Sharma, S.C.: Composite Materials. Narosa Publishing House, New Delhi (2000)
5. Harris, B.: Engineering composite materials, IOM (1999)
6. Gay, D.: Composite Materials: Design and Applications. CRC Press, Boca Raton (2014)
7. Strong, A. B.: Fundamentals of composites manufacturing: materials, methods and applications. Society of Manufacturing Engineers (2007)
8. Shalin, R.E.: Polymer Matrix Composites. Chapman and Hall, Boca Raton (1995)
9. Wang, R.M., Zheng, S.R., Zheng, Y.G.: Polymer Matrix Composites and Technology. Woodhead publishing in materials, Cambridge (2011)
10. Hunt, J., B., Howes, J. B., Hunt, G.: Waste reduction programs for commercial/industrial solid waste. <http://citeseerx.ist.psu.edu/viewdoc/download?doi=10.1.1.399.2000&rep=rep1&type=pdf>
11. Ashori, A.: Hybrid composites from waste materials. J Polym Environ **18**, 65–70 (2010)
12. Medupin, R.O., Abubakre, O.K., Ukoba, O., Imoisili, P.E.: Mechanical properties of wood waste reinforced polymer matrix composites. Am. Chem. Sci. J. **3**(4), 507–513 (2013)
13. Koleva, M., Zheglova, A., Fidancevska, E., Vassilev, V.: Composites Containing Waste Materials. INTECH Open Access Publisher, Croatia 2011
14. Sathiyamoorthy, M., Thanappan, S., Senthilkumar, M.: Utilization of waste rubber tires as an additional ingredient of concrete mixtures. Int. J. Eng. Res. Ind. Appl. **4**(I), 335–350 (2011)
15. Tunsan, M.: The use of waste materials in asphalt concrete mixtures. Waste Manag. Res. **21**(2), 83–92 (2003)
16. Vassilev, V., Fidancevska, E., Milosevski, M., Parvanov, S., Milosevski, D., Hristova-Vasileva, T.: Composites based on industrial wastes IV. Production of porous composites from Fe-Ni slag and waste glass. J. Univ. Chem. Technol. Metall. **42**(4), 369–376 (2007)
17. Thomas, C., Borges, P.H.R., Panzera, T.H., Cimentada, A., Lombillo, I.: Epoxy composites containing CFRP powder wastes. Compos. B **59**, 260–268 (2014)
18. Andoglu, E.M., Akpinar Borazan, A., G. Acikbas: Investigation of mechanical properties of polyester/marble dust and hop wastes reinforced composite material, 14th International conference on environmental science and technology. http://cest.gnest.org/cest15proceedings/public_html/papers/cest2015_00391_poster_paper.pdf
19. Gokdai, D., Akpinar Borazan, A., Acikbas, G.: Effect of marble: pine cone waste ratios on mechanical properties of polyester matrix composites. 14th International conference on environmental science and technology, Rodos. http://cest.gnest.org/cest15proceedings/public_html/papers/cest2015_00519_oral_paper.pdf
20. Alok, S., Amar, P.: Analysis of dry sliding wear behaviour of red mud filled polyethylene composites using taguchi method". J. Reinf. Plast. Compos. **29**(24), 1–15 (2008)
21. Parvaiz, M.R., Mohanthy, S., Nayak, S.K., Mahanwar, P.A.: Polyetherketone: composites reinforced with fly ash and mica. J. Miner. Mater. Charact. Eng. **9**, 25–41 (2010)

22. Biswas, S., Satapathy, A., Patnaik, A.: Effect of ceramic fillers on mechanical properties of bamboo fiber reinforced epoxy composites: a comparative study. *Adv. Mater. Res.* **123–125**, 1031–1034 (2010)
23. Kumar, N.V., Sarcar, M.M.M., Ramji, K., Geetha, V.: Evaluation of wear properties of industrial waste (slag) reinforced polypropylene composites. *Int. J. Theor. Appl. Res. Mech. Eng.* **3**, 2319–3182 (2014)
24. Mishra, P., Bera, T.: Fabrication and characterization of composite using carbon black-waste from petroleum products. *Int. J. Adv. Res.* **2**(7), 122–130 (2014)
25. Agunsoye, J.O., Talabi, S.I., Obidiegwu, E.O., Awe, I.O.: The development and characterization of epoxy-foundry basic slag particulate composite materials. *Int. J. Compos. Mater.* **4**(2), 93–102 (2014)
26. Acikbas, G., Acikbas Calis N., İkizek, E., Özel, M., Eker, A.S.: Characterization of green epoxy matrix composites filled with ceramic wastes. *ISITES2014 2nd International symposium on innovative technologies in engineering and science proceeding book*, pp. 597–606, Karabük, Turkey, 18–20 June 2014
27. Biswas, S., Satapathy, A., Patnaik, A.: Effect of ceramic fillers on mechanical properties of bamboo fiber reinforced epoxy composites: a comparative study. *Adv. Mater. Res.* **123–125**, 1031–1034 (2010)
28. Barbuta, M., Rujanu, M., Nicuta, A.: Characterization of polymer concrete with different wastes additions. *Proced. Technol.* **22**, 407–412 (2016)
29. Fombuena, V., Bernardi, L., Fenollar, O., Boronat, T., Balart, R.: Characterization of green composites from biobased epoxy matrices and bio-fillers derived from seashell wastes. *Mater. Des.* **57**, 168–174 (2014)
30. Goh, C.K., Valavan, S.E., Low, T.K., Tang, L.H.: Effects of different surface modification and contents on municipal solid waste incineration fly ash/epoxy composites. *Waste Manag.* (2016). doi:[10.1016/j.wasman.2016.05.027](https://doi.org/10.1016/j.wasman.2016.05.027)
31. Purohit, A., Satapathy A.: Epoxy composites filled with micro-sized LD sludge: wear characterization and analysis. 5th National conference on processing and characterization of materials proceeding book, vol. 115, pp. 1–8, (2016)
32. Prithvirajan, R., Jayabal, S., Bharathiraja, G.: Bio-based composites from waste agricultural residues: mechanical and morphological properties. *Cellul. Chem. Technol.* **49**(1), 65–68 (2015)
33. Martins, A.L.B.S., Gouvea, R.A., Oliveria, M.P., Candido, S.N.: Characterization of epoxy matrix composites incorporated with sugarcane bagasse fibers. *Mater. Sci. Forum* **775–776**, 102–106 (2014)
34. PickeringS, J.: Recycling technologies for thermoset composite materials-current status. *Compos. A* **37**, 1206–1215 (2006)
35. Alfred R. Paz, Bound Brook, N. J., Process of preparing urea-formaldehyde molding composition, UP2,872,425 (1959)
36. Sims, G., Bishop, G: UK polymer composites sector-competitive analysis and foresight study. Final Report, Published by NPL Materials Centre, July 2001
37. Barari, B., Omrani, E., Moghadam, A.D., Menezes, P.L., Pillai, K.M., Rohtagi, P.K.: Mechanical, physical and tribological characterization of nano-cellulose fibers reinforced bio-epoxy composites: an attempt to fabricate and scale the “Green” composite. *Carbohydr. Polym.* **147**, 282–293 (2016)
38. Borsoi, C., Zattera, A.J., Ferreira, C.A.: Effect of cellulose nanowhiskers functionalization with polyaniline for epoxy coatings. *Appl. Surf. Sci.* **364**, 124–132 (2016)
39. Economic Importance, Formaldehyde Council 2009. Accessed 14 April 2010
40. Acikbas, G.: Development and characterization of polymer matrix composite materials alternative to ceramic sanitarywares. *Mater. Sci. Eng., Ph.D. Thesis*, 2016, Dumlupinar University, Turkey
41. Prasada, A., Fotoua, G., Lia, S.: The effect of polymer hardness, pore size, and porosity on the performance of thermoplastic polyurethane-based chemical mechanical polishing pads. *J. Mater. Res.* **28**(17), 2380–2393 (2013)