SYNERGISTIC EFFECT OF HYBRID REINFORCEMENTS ON MECHANICAL AND THERMAL PROPERTIES OF EPOXY COMPOSITES FOR INSULATION MATERIALS – A COMPARATIVE STUDY

MURUGAN VIGNESH and MUTHU PARANTHAMAN VENKATESH

Department of Civil and Structural Engineering, Annamalai University, Annamalainagar 608002, India
✉ *Corresponding author: M. P. Venkatesh, ermpvenk@gmail.com*

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The growing concerns regarding environmental sustainability have catalysed the exploration and adoption of sustainable materials across various industries. Polymer composites have emerged as a significant contender in this arena. This research delves into the thermal and mechanical characteristics of epoxy matrix composites reinforced with micron-sized waste paper fillers and hybrid fillers. Utilising the hand lay-up technique, composites were meticulously fabricated with sustainable micro-filler and natural fibre ensuring their uniform dispersion within the epoxy matrix. A comprehensive mechanical assessment of these composites was performed, juxtaposing them against the mechanical properties of the pristine epoxy. This evaluation encompassed tensile, flexural, and hardness tests. Interestingly, an optimal enhancement in thermal and mechanical properties was found at the peak filler concentration.

Keywords: sustainable materials, polymer composites, epoxy matrix, waste paper fillers, mechanical properties, construction

INTRODUCTION

To improve energy efficiency, inorganic thermal insulators are often utilised. However, environmental and health concerns about their manufacture and up to the end of life have grown.[1] As time and technology have progressed, various natural materials have been utilised as an alternative and supporting element in the development of insulators. Renewable resources, such as natural fibres (hemp, coir, flax, *etc*.), bagasse and sawdust, are a few of them. For instance, bagasse is the fibrous residue obtained in the sugarcane milling plant after extracting the juice from the sugarcane stalks. Bagasse has a lot of valuable components that can be reprocessed to develop composite materials.[2] For example, Usubharatana *et al.* used bagasse and other agricultural waste to develop thermal insulators.[3] The authors analysed the thermal conductivity and conducted a life cycle assessment (LCA) of four renewable source-based thermally insulating materials. The four agricultural waste thermal insulators analysed had thermal conductivities ranging from 0.042 to 0.087 W/mK. Also, the thermal conductivity of the materials made with concentrated latex chemicals was found to be the lowest, while the materials made by hot-pressing had the greatest. As regards the life cycle assessment, the rice straw insulator had a significant environmental impact, followed by the materials made from bagasse, coconut coir, and oil palm fibre, in that order. The eco-efficiency of an oil palm fibre insulator made from combined concentrated latex chemicals was the best, even compared to commercial thermal insulators.[3]

In the development of foam based insulators, the use of technical lignin has been investigated. However, because of its low compatibility with other polymers, lignin application for this purpose is not straightforward. Several process steps, such as adjusting the molar mass, separating sugars, extractives, and inorganic compounds, focusing on modification reactions for reactivity advancement via implementation of novel active sites, functionalizing hydroxyl groups, or copolymerizing with other suitable polymers, could be used to solve the issue.[4]

For good thermal insulators, low thermal conductivity (K) and good durability are important characteristics.[5] Bruijn *et al*. found that adding hemp fiber to a composite formulation gave it better thermal qualities, including decreased thermal conductivity and specific heat capacity.[6] Also, cork particles can be used as a good insulator, the chemical, physical, and mechanical properties of cork have been demonstrated.[7] The value of K, thermal diffusivity (α), and heat capacity (c) of granulated cork based composite materials were explored by Chekri *et al.* In the steady state regime, the thermal conductivity is determined using an alternate hot plate approach. The results demonstrated that using particles in the granular class D = 6.3–8 mm and granular cork content larger than 40%, a homogeneous granular composite material could be obtained.[7] The feasibility of employing cork–gypsum composites was investigated by Olivares *et al.*[8] and Chekri *et al.*[9] The study of Olivares *et al*. demonstrated that the obtained cork-gypsum composites have the potential to be employed as partition walls due to the thermal insulation provided by the cork.[8] Furthermore, the findings of Chekri *et al*. demonstrated the efficacy of the asymmetrical transient hot plate approach for thermal assessment of high-insulating granular materials.[9]

Some research works reported the utilisation of waste paper in polymer composites using high-density polyethylene as matrix.[10] Moreover, Arhab *et al.* manufactured a novel, sustainable insulation material made with a hybrid filler combining recycled paper waste and *Ampelodesmos mauritanicus* fibers. The developed insulator offered excellent thermal insulation (0.027 W/m.K), durability, and cost-effectiveness, promoting eco-friendly development practices.[11]

Considering that environmentally friendly practices should involve the recycling of wastes and use of renewable resources,[12] this study aimed to develop effective thermal insulators for construction applications. The insulating materials were prepared by incorporating varying weight percentages of waste paper particles into an epoxy matrix to create composites. Also, to avoid agglomeration issues that appeared with higher contents of paper particles, the filler was hybridised with betel nut fibre. The experimental findings demonstrated notable improvements in mechanical properties.

EXPERIMENTAL

Materials and their preparation

The polymeric matrix used in this study was an epoxy resin, with the density of 1.42 g/c^3, and a corresponding hardener, with the density of 1.22 g/c^3. The epoxy is a thermoset type of polymer that is known to be used in various microelectronic, structural and automotive applications.

For this study, waste papers were collected from a local shop, and paper particles were obtained through a step-by-step size reduction process. First, the waste paper was torn into small pieces and immersed into water for 48 h. Further, the wet paper was agitated by hand to help it disintegrate. The disintegrated paper was removed from the water and allowed to dry. Then, it was converted into powder using a flour mill.

Betel nuts were obtained from local farms in Tamil Nadu, and soaked in water for 18–24 hours to facilitate gentle extraction of the fibers. The fibers were manually extracted to avoid damage, then were sun-dried for 48 hours to eliminate moisture. The dry fibers were stored in a container in order to maintain fiber integrity until further composite preparations.

Composites preparation procedure

The epoxy resin and the hardener were mixed together to prepare the matrix material. The epoxy-to-hardener ratio was set at 10:1, in accordance with previous research.[13,14]

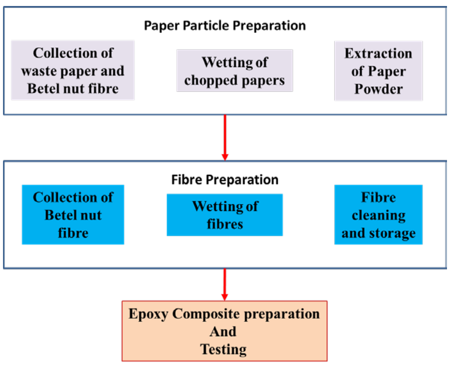


Figure 1: Preparation procedure of the reinforced epoxy composites

Table 1
Formulations of epoxy based composites and corresponding sample codes

Matrix	Reinforcement (wt%)		Sample code
	Paper particles	Betel nut fibres	
Epoxy	0	0	Neat epoxy
	2	0	2P wt%
	4	0	4P wt%
	6	0	6P wt%
	1	1	1P + 1B wt%
	2	2	2P + 2B wt%
	3	3	3P + 3B wt%

Figure 2: Epoxy based composite samples

Two sets of samples, with different formulations, were prepared by the hand lay-up method. In the first set, paper particles were used as reinforcement to produce the composites, in varying loadings (2, 4, 6 wt%).

The paper particles were uniformly mixed with the epoxy matrix, poured into molds, and allowed to cure at room temperature.

In the other set, hybrid composites were prepared, using combined fillers: paper particles and betel nut fiber, as reinforcements, in varying loadings (1, 2, 3 wt% for each filler). The fillers were blended with the epoxy matrix, cast into molds, and cured under identical conditions. In addition, a pure epoxy material was also prepared in a similar manner.

After the curing time, all the composites were demolded, trimmed, and prepared for subsequent testing and analysis. Figure 1 shows the steps of the composite preparation procedure and Table 1 lists the detailed formulations of all the composites and their corresponding sample codes.

Testing and characterization

Tensile and flexural tests

In this study, mechanical characteristics were assessed through tensile and flexural testing. Each category involved testing three composite samples, and average tensile strength and elongation values were recorded. Tensile and flexural properties of the epoxy composite specimens (Fig. 2) were evaluated by tensile and three-point bend tests, respectively, conducted on a Tinius Olsen Universal Testing Machine (UTM), at room temperature, with a constant strain rate of 1 mm/min. Specimens were prepared according to ASTM D638-10 and ASTM D790-10 standards for tensile and flexural tests, respectively.

Thermal conductivity

The thermal conductivity of the prepared epoxy based composites was measured using a Laser Flash apparatus. The transient plane source (TPS) approach is one of the most accurate and simple ways of researching thermal transport parameters that provides details on the material properties, such as K, α and c values.

RESULTS AND DISCUSSION

Tensile properties

The tensile properties of the composite materials, reinforced with a single or hybrid fillers, in various loadings, were examined. The tensile testing revealed that the incorporation of both paper particles and betel nut fibres as hybrid fillers in the epoxy based composites enhanced tensile strength more effectively than using paper particles alone. It was noted that, while the tensile strength of the epoxy matrix material was 22.6 MPa, that of the epoxy composites reinforced with 2P wt%, 4P wt%, and 6P wt% paper particles showed increases of 5%, 17%, and 24% in tensile strength, respectively, compared to the reference epoxy (Fig. 3). This enhancement is attributed an efficient stress transfer between the epoxy and the paper particles.

However, the agglomeration of paper particles is a limiting factor to be considered. It was noticed that an increasing content of the waste paper particle reinforcement caused issues such as agglomeration and uneven dispersion of the filler within the matrix, therefore, the loading of this filler was limited to 6 wt% during composite fabrication.

To address these issues, hybrid epoxy composites incorporating both paper particles and betel nut fibres were developed. These hybrids outperform the materials with only paper particle reinforcement in tensile strength. Specifically, 1P+1B wt%, 2P+2B wt%, and 3P+3B wt% hybrid fillers reinforced epoxy composites exhibited improvements of 14%, 22%, and 38% in tensile strength, respectively, compared to the reference epoxy. The primary contributor to this increase is the load-sharing capacity of natural fibres, with betel nut fibres aiding the epoxy in sustaining higher loads.

Epoxy composites generally exhibit lower elongation at break compared to the base epoxy resin, highlighting the influence of the reinforcing phase on the material's deformation behaviour. The consistent decrease in elongation with increasing loadings of the fillers in the composite fabrication suggests a growing brittleness of the resulting composites. However, the hybrid filler-reinforced epoxy composites demonstrate superior elongation at break, compared to the material reinforced with paper particles alone.

This improvement in elongation can be attributed to the bonding between betel nut fibres and the epoxy, allowing the composite specimens to endure more deformation. It seems like the combination of paper particles and betel nut fibres not only enhances tensile strength, but also provides better flexibility, mitigating the overall brittleness of the composites. This suggests a promising potential for the hybrid filler reinforcement in enhancing the mechanical properties of epoxy based composites.

The fractured morphology of the composite materials was examined and SEM images are illustrated in Figure 4, strongly suggesting brittle failure. Moreover, numerous pores are visible within the epoxy composites reinforced exclusively with paper particles. The betel nut fibres demonstrate sufficient bonding with the epoxy matrix to provide better tensile strength, the images suggest that an enhanced interfacial bonding could yield even greater improvements in mechanical properties.

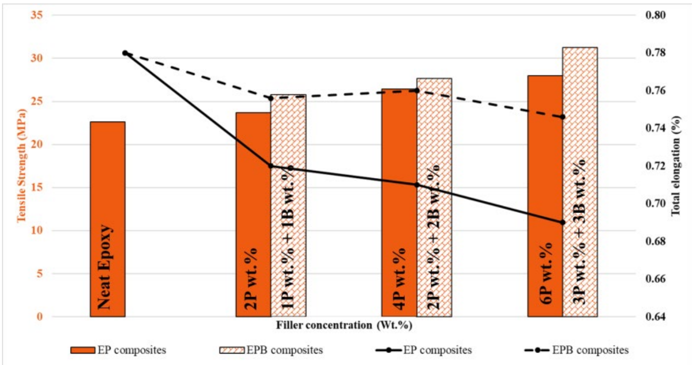
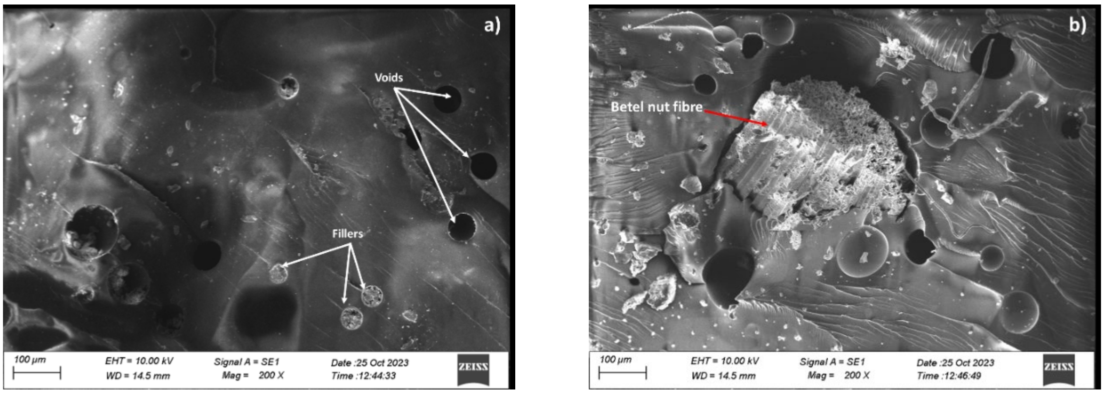


Figure 3: Tensile properties of epoxy based composites reinforced with single or hybrid fillers



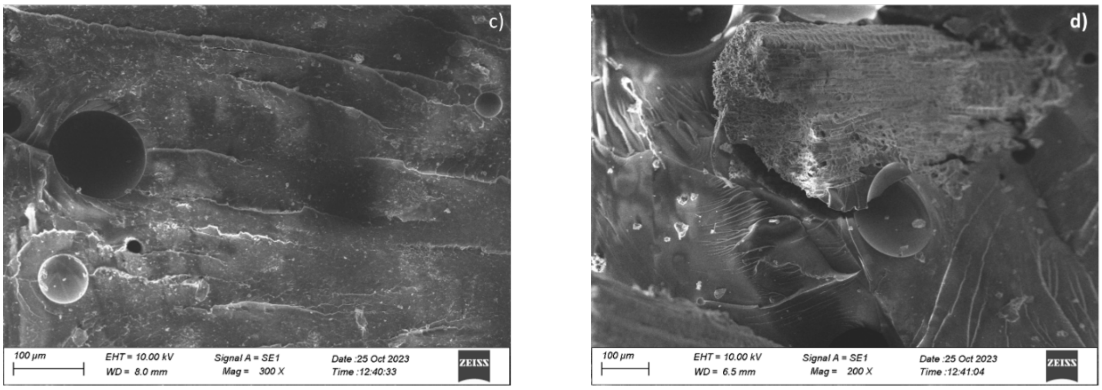


Figure 4: Morphology of fractured surfaces during tensile tests for (a) 4P wt%, (b) 2P+2B wt%, (c) 6P wt% and (d) 3P+3B wt% composites

Flexural properties

The study involves testing and comparing the flexural properties, namely flexural strength and elongation at break, of the epoxy based composites reinforced solely with paper particles and those with hybrid reinforcement, compared with that of the base epoxy. Interestingly, an increasing trend of flexural strength was recorded with increasing filler loading, especially for the hybrid fillers, similarly to the trend of the tensile strength results.

The hybrid filler-reinforced epoxy composites demonstrated superior performance, compared to the epoxy composites reinforced solely with paper particles. Thus, the neat epoxy exhibited a flexural strength of 35.6 MPa, and the addition of 2 wt%, 4 wt%, and 6 wt% of paper particles led to improvements of 9%, 13%, and 19%, respectively (Fig. 5). As regards the hybrid fillers, the 1P+1B wt%, 2P+2B wt%, and 3P+3B wt% reinforced epoxy composites showcased even more substantial enhancements, with improvements of 16%, 23%, and 22%, respectively, in flexural strength (Fig. 5).

It is intriguing to observe the notable advantages that the hybrid filler combination brings to the flexural properties of the epoxy composites. The elongation at the break for all epoxy composite combinations is lower than that of the base epoxy, attributed to the increased brittleness with added reinforcements. Interestingly, the hybrid filler-reinforced epoxy composites exhibit superior elongation at break compared to epoxy composites filled with paper particles alone, across all filler concentration levels. The observed decrease in elongation at break suggests a shift towards increased brittleness upon reinforcement.

Overall, hybrid reinforced epoxy composites demonstrate superior tensile and flexural strength, compared to those reinforced with a single filler – of paper particles. This enhancement is attributed to the excellent load-bearing capacity of betel fibre, which, being continuous, ensures efficient stress transfer across the matrix. The cellulose present in betel fibres contributes to strong interfacial bonding with the epoxy resin, reducing voids and improving mechanical performance. In contrast, the discontinuous nature of paper particles and their relatively lower adhesion with the matrix limit their ability to reinforce effectively. The presence of voids in the composite, as evident in the FESEM images, is attributed to the manual hand-stirring process used during mixing. This issue can be mitigated by employing speed-controlled mechanical stirring or vacuum-assisted stirring, both of which promote more uniform mixing and effectively minimise air entrapment within the material.

Shore D hardness

The Shore D hardness of polymers can vary depending on the specific formulation and curing method. The epoxy resins typically exhibit Shore D hardness values in the range of 80 to 82, as confirmed by the shore D hardness test results in this study (Fig. 6). The Shore D hardness of epoxy composites experienced slight enhancement with the inclusion of paper particles reinforcement. For instance, the 6P wt% paper particle-reinforced epoxy composite demonstrated a 6% increase in Shore D hardness, compared to the base epoxy. Meanwhile, the 3P+3B wt% hybrid epoxy composite exhibited a notable 9% rise in Shore D hardness. This improvement is primarily attributed to the incorporation of betel nut fibre. Other composite manufacturing methods, such as compression moulding, could potentially allow achieving further increases in hardness.

Thermal conductivity

The thermal conductivity of the neat epoxy sample and its composites is presented in Figure 7. In contrast to the neat epoxy, all other epoxy composite samples showed a noticeable decrease in thermal conductivity, primarily attributed to reduced crystallinity and enhanced crosslinking. Including reinforcements, as paper particles and betel nut fibres in this case, impeded the mobility of the epoxy during curing, leading to improved polymer crosslinking.

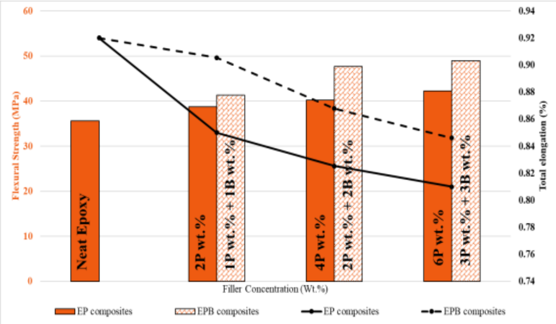


Figure 5: Flexural properties of epoxy-based composites

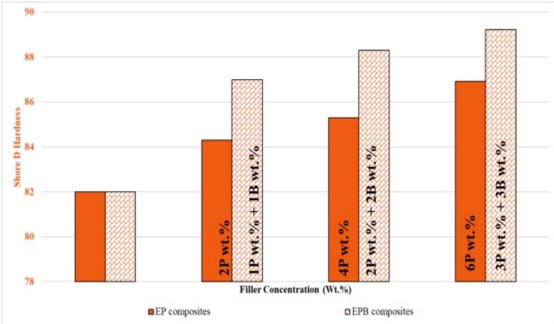


Figure 6: Shore D hardness of epoxy-based composites

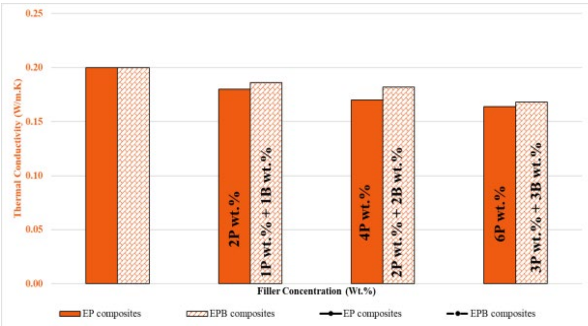


Figure 7: Thermal conductivity of epoxy-based composites

Moreover, the presence of porosity in the epoxy composites during fabrication also contributes to decreased thermal conductivity. The combination of natural fibres and fillers consistently exhibits decreased thermal conductivity across all concentrations of the filler reinforcement, demonstrating the efficacy of the developed materials as a thermal insulator in construction applications.

CONCLUSION

Epoxy based composites, with paper particle reinforcement and hybrid paper particle and betel nut fibre reinforcement, were prepared using the hand lay-up method. The mechanical properties and thermal performance of the developed materials were assessed. From the mechanical and thermal testing, the conclusions below could be drawn.

The tensile strength of the epoxy composites rose with higher concentrations of the paper particles filler, and this enhancement was augmented by the introduction of betel nut fibres into the epoxy based composites, as part of a hybrid reinforcement. However, the elongation of the composites was reduced with the incorporation of reinforcements. In addition, a rising trend in flexural strength was observed, mirroring the results seen in tensile strength. The epoxy composites reinforced with hybrid fillers exhibited superior performance when compared to those reinforced solely with paper particles. Likely, the hardness test revealed an increase in the Shore D value of the epoxy composites. Further, the thermal conductivity of both types of epoxy composites showed a lower value than that of the neat epoxy, indicating their better insulation capability.

Overall, the hybrid reinforcement (consisting of both fibres and particles) contributed to better mechanical and thermal performance. However, voids and agglomeration of the fillers are the major hindrances, which could be addressed through the selection of more advanced manufacturing methods. This could yield novel epoxy based composite materials with promising mechanical properties and thermal performance for indoor thermal insulation applications.

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