Rice husk and saw dust reinforced hybridized blends of bio-benzoxazine/epoxy based composites: Thermal, Mechanical, and Acoustic Absorption Properties

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Research Article

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Abstract

The prime objective of the present work is to develop bio-wastes (rice husk and saw dust) reinforced composites with varying weight percentages composition of hybridized blend made from bio-based cardanol-melamine, cardanol-aniline benzoxazine and epoxy resin (bio-benzoxazine : epoxy resin 0:100 and 50:50 wt%) for acoustic proof applications. The breakthrough achieved in the present work is the cardanol benzoxazine is made in the form of hybrid matrix with epoxy resin, which cures significantly at lower temperature of 105°C in the absence of any curatives and also cures at room temperature in the presence of isophorone diamine curative, whereas the cardanol-melamine and cardanol-aniline based benzoxazines cure at very high temperature of 221°C and 275 °C respectively. The low temperature cure behaviour achieved in the present work facilitates the amenable fabrication of bio-composites using both low heat resistant bio-based reinforcements and high temperature cure bio-benzoxazines for making cost competitive green building materials. Cardanol-melamine (C-m) and cardanol-aniline (C-a) based benzoxazines were synthesised using cardanol separately with melamine (m) and aniline (a) in the presence of paraformaldehyde under suitable experimental conditions. The benzoxazine (C-m and C-a) obtained was blended with varying weight percentages of bisphenol-A epoxy (DGEBA) resin separately followed by reinforcing with rice husk and saw dust and then cured at room temperature with stoichiometric quantity of isophorone diamine to obtain corresponding hybrid bio-composites. Mechanical properties (tensile strength, modulus, percentage elongation and hardness), thermal conductivity, thermal resistance, and sound absorption coefficient were studied as per standard methods. Results obtained from different studies infer that hybrid blended cardanol benzoxazine composite panels reinforced with rice husk and saw dust possess appreciable thermal, mechanical and acoustic properties. Data obtained from acoustic studies, it was observed that the highest value of sound absorption coefficient of 6400 Hz was noticed for composite specimens developed using both rice husk and saw dust reinforced with hybridized blend of bio-benzoxazine (50wt%) and epoxy resin (50wt%) and these hybrid composite panels can be used as sound absorption material in the ceiling and wall construction applications.

Introduction

The rapid industrialization and urbanization contribute to undesired sound pollution to the environment and is called noise pollution and it causes lot of problems to the environmental quality and human, hence its reduction or prevention is essential to protect human health and maintaining the quality of life. The acoustic proof materials are used in the construction of floors, walls and ceiling in the field of construction of buildings (Asdrubali et al. 2017), auditoriums (Elkhateeb 2012), theatres (Chourmouziadou and Kang 2008), automobiles (Siano et al. 2016), aerospace (Paun et al. 2003) and other transport systems to alleviate problems associated with noise pollution.

The use of acoustic panel is one of the most important methods of practice to provide sound insulation in the field of fabrication of equipments and construction of automobiles and buildings. (Chen et al. 2019) Acoustic absorption panels made from natural fibres are less hazardous to human health and more eco-
friendly than those made of conventional synthetic fibres. (Mamtaz et al. 2016; Bhingare et al. 2019; Liuzzi et al. 2020; Tang and Yan 2020) (Balčiūnas et al. 2016) Hence, to satisfy environmental issues, synthetic materials need to be replaced with suitable natural fibres. Natural waste have been widely used due to their inherent properties such as bio-degradability, renewability and their abundant availability, in addition to their light weight, carbon neutral and cost competitiveness. (Thyavihalli Girijappa et al. 2019) (Guna et al. 2019)

It was reported that the natural fibrous materials compete with synthetic materials with effective sound absorbing behaviour viz., barley straw, (Liuzzi et al. 2020) flux, (Sair et al. 2019) cotton, (He et al. 2018) saw dust, (Bansod et al. 2016) rice husk, (Taban et al. 2019) hemp, (Liao et al. 2020) palm, (Mohammed et al. 2018) (Belakroum et al. 2017) kenaf, (Ismail et al. 2019) corn, (Sari et al. 2017) (Binici et al. 2016) paddy, (Marques et al. 2020) sisal, (Tholkappiyana et al. 2015) banana, (Singh and Mukhopadhyay 2020) and cork. (Trematerra and Lombardi 2017) Further, natural fiber reinforced polymer composites have also received a lot of attention due to their lightweight, bio-degradable nature, eco-friendly nature. Though the natural fibres possess a number of advantages, some of their short comings like low interfacial adhesion, poor moisture resistance, and inferior microbial resistance need to be alleviated in order to utilize them towards effective sound absorption applications.

In the recent years, researchers turned their attention towards the sustainable and renewable bio-based polymeric matrices to develop composites for different industrial applications including sound absorption uses. In this context, the most probable matrix resin suitable for the fabrication of composites is considered as polybenzoxazines, since these resins possesses easy process-ability, cures without release of by-product, low shrinkage behaviour, molecular flexibility, excellent thermal stability, improved chemical resistance, good mechanical properties, excellent hydrophobic nature and cost competitive. (Hariharan et al. 2017a, 2018) (Kim and Ishida 2001; Froimowicz et al. 2016; Kotzebue et al. 2018; Lyu and Ishida 2019) (Muthukaruppan et al. 2018) Till date, relatively, a meagre attention has been paid to bio-based benzoxazine composites. (Wang et al. 2012) In the recent past researchers directed their attention towards bio-based polybenzoxazines in particular cardanol based benzoxazines and their applications. (Arumugam et al. 2018; Hariharan 2020) Cardanol based benzoxazines are versatile bio-based polymeric matrices suitable for fabrication of reinforced composites. (Shukla et al. 2017) Since, they possess good hydrophobic nature due to the presence of m-substituted long aliphatic chain and it contributes to reduce the brittleness as well as enhances the hydrophobic behaviour. Recently, our research group have reported benzoxazine composites based on cardanol for low dielectric, (Hariharan et al. 2019) oil-water separation (Hariharan 2020) and corrosion resistance applications. (Lakshmikandhan et al. 2019; Selvaraj et al. 2019; Hariharan et al. 2020)

In the present work an attempt has been made to develop composites based on bio-wastes (rice husk and saw dust) reinforced using hybrid matrices blended with cardanol benzoxazine and bisphenol-A epoxy resin in order to utilize them for acoustic absorption applications. In this context, cardanol-melamine (C-m) cardanol-aniline (C-a) based benzoxazines were synthesised using melamine, aniline
and paraformaldehyde separately. The biobased benzoxazines synthesized were then blended with varying weight percentages of bisphenol-A epoxy resin (DGEBA) and reinforced separately with rice husk and saw dust to obtain corresponding bio-composites. The specimens of composites prepared have been characterized for their physico-chemical, thermal, mechanical and acoustic insulation behaviour by different analytical methods and the results obtained are discussed and reported.

The importance of the present work is utilizing high temperature curable (about 221°C) bio-benzoxazines made compatible with less heat resistant natural fibres (rice husk and saw dust) for developing hybrid composites with epoxy resin and isophorone diamine curable at room temperature to facilitates the ease of processing and fabrication in order to save energy, time, cost and to exploit the benefits of both bio-benzoxazine resin and bio wastes. The importance of the present work is considered as the most of the materials used are either from sustainable bio-source or commercially available cost competitive materials. To the best our knowledge, no reports are available as on date about the hybridized blend of bio waste reinforced bio-resin based composites. The present work is considered as the first its kind to use blended hybrid cardanol-benzoxazine/epoxy resin matrix reinforced with rice husk and saw dust in the form of sound proof composite panels.

Experimental

Materials

Cardanol was obtained from Satya Cashew Products, Chennai. The bisphenol-A epoxy resin (DGEBA) was purchased from Roto Polymers Ltd, Chennai, India. Rice husk and saw dust were collected from Coimbatore, India. Paraformaldehyde and anhydrous sodium sulphate (Na₂SO₄), melamine and aniline were obtained from Sigma-Aldrich, India. Ethyl acetate and sodium hydroxide were obtained from SRL, India.

Synthesis Of The Cardanol-aniline Benzoxazine (c-a)

Cardanol based benzoxazine was synthesised using aniline and paraformaldehyde using previously reported procedure. 1 mol. of bio-phenol (cardanol-C), 1 mol. of aniline (a) and 2 mol. of paraformaldehyde were placed in a one litre three necked round bottomed flask equipped with mechanical stirrer and thermometer. The reaction was conducted by heating with continuous agitation at 100°C for 5h in the absence of any solvents. The progress of the reaction was monitored by thin layer chromatography. The obtained reaction product was diluted by the addition of ethyl acetate solvent to remove unreacted material present, if any, and was thoroughly washed with distilled water using a separating funnel. Then, the organic phase obtained was collected and dried with anhydrous sodium sulphate and filtered. The solvent left in the product was removed under vacuum.(Arumugam et al. 2018)

Synthesis Of The Cardanol-melamine Benzoxazine (c-m)

0.3 mol of bio-phenol (cardanol-C), 0.1mol of melamine (m) and 0.6mol of paraformaldehyde were placed in a 500 ml three necked round bottomed flask equipped with magnetic stirrer, thermometer and
reflux condenser. The reaction mixture was heated under stirring at 100°C for 5 h. The crude of the reaction (Scheme 3), product obtained was diluted with the addition of ethyl acetate and was thoroughly washed using distilled water using a separating funnel. Then, the organic phase obtained was collected and dried with anhydrous sodium sulfate and filtered. The solvent left in the product was removed under vacuum. The product benzoxazine was isolated (Scheme 3) and characterized by $^1$H NMR and FT-IR.

**Treatment of rice husk and saw dust.**

The husk and saw dust obtained were heated separately with 10% NaOH solution at a temperature of 80°C for 1 h. to remove non-cellulosic impurities. After the chemical treatment, the fibres were washed repeatedly with distilled water, and then dried at 60°C for 24 h (Fig. 1). The alkali treatment, enhances porosity and higher surface area by providing a more tortuous path and in turn improves the low frequency sound absorption and removes air flow resistivity of fibrous materials. The increase of air flow resistivity causes loss of sound energy through friction of sound waves with air molecules and thus improves low frequency sound absorption. The present work studies the limitations of husk and saw dust to assess their acoustic absorption performance at a desired level. Further, thermal conductivity and acoustic insulation behaviour of natural fibres reinforced bio-benzoxazine/epoxy composites were studied and discussed.

**Preparation Of Neat And Bio-waste Reinforced Composites**

The curing temperature observed for cardanol benzoxazines (C-m, C-a) were considered to be very high (221 and 275°C). However, in the case of blend of bis-phenol-A epoxy (DGEBA) and cardanol benzoxazine (C-a, C-m), the curing temperature was found to be around 100°C in the absence of any curative. Furthermore, it was also ascertained that the benzoxazine/epoxy blend can be cured at room temperature using isophorone diamine as curative for the blends to facilitate preparation of bio-composites. The 50wt%:50wt% of (C-a, C-m) bio-benzoxazine/bisphenol-A epoxy (DGEBA-isophorone diamine) matrix panel specimen was made without any reinforcement (Schemes 1 and 2). The chemically treated and dried bio-wastes were separately reinforced with bio-benzoxazine/epoxy to obtain composite panels (Fig. 1 and Scheme 3). 100g of chemically treated rice husk and saw dust were separately soaked and reinforced with 200g of neat DGEBA-isophorone diamine blend followed by mixed 50:50 wt% of C-a /DGEBA-isophorone diamine and C-m /DGEBA-isophorone diamine and then separately poured into a mould of 300 × 300 × 6 mm and cured for overnight at room temperature under mild pressure to obtain corresponding composites. The composite samples obtained were utilised for further studies.

**Measurements**

FTIR spectra measurements were carried out with Agilent Cary 630 FTIR Spectrometer. NMR spectra were obtained with Bruker (400 MHz) using deuterated chloroform (CDCl$_3$) as a solvent and tetramethylsilane (TMS) as an internal standard. DSC measurements were recorded using NETZSCH STA 449F3 under N$_2$ purge (60 mL min$^{-1}$) at heating rate of 10 °C min$^{-1}$. The thermal conductivity analyses were carried out
using heat flow meter. Sound absorption (acoustic insulation behaviour) was determined using the impedance tube method. Mechanical behaviour of neat and composite samples were analysed using INSTRON 8801 as per ASTM standards.

**Results And Discussion**

The cardanol based benzoxazine was synthesized using cardanol, amine (melamine or aniline) and paraformaldehyde through Mannich condensation reaction in the absence any solvent. After the appropriate work-up, cardanol-aniline (C-a) and cardanol-melamine (C-m) benzoxazines were extracted and characterized. The molecular structure of benzoxazines were confirmed from FT-IR and $^1$H NMR spectral analysis. The curing behaviour of benzoxazine as well as blend of benzoxazine/bisphenol-A epoxy (DGEBA) was studied by DSC analysis.

**Curing Behaviour**

The occurrence of thermal polymerisation of C-a, C-m benzoxazine and benzoxazine/epoxy (DGEBA) blends was studied using DSC analysis and the results obtained are presented in Fig. 3. The appearance of exothermic peak confirms the occurrence polymerization through thermal ring-opening mechanism. The homo polymerization temperature (Tp) of benzoxazine (C-a) was observed at 275°C from the appearance of exothermic peak (Fig. 2), which is significantly higher than that of conventional benzoxazines, normally they cure over the range of temperature between 220°C and 260°C. (Hariharan et al. 2017b; Ručigaj et al. 2015; Zeng et al. 2019; Zhan et al. 2020) The homo polymerisation of DGEBA is observed at 390°C (Figure S1) in the absence of any catalyst/curatives.

The appearance of exothermic peak in the differential scanning calorimetry (DSC) curve can be used to predict the temperature conditions required for polymerization (Tp) of benzoxazine/epoxy blends. The thermal-induced homo polymerization of epoxy was much higher when compared with that of curing with appropriate curing agent, which facilitates the occurrence of curing reaction at room temperature. This will led to the facile processing of composites at room temperature which will reduce the consumption of energy and in turn lowers the cost of production. This is the one of the major advantages of the present work by hybridizing both bio-based benzoxazine and synthetic epoxy resin to obtain balanced properties suitable for the development of hybrid composites. The polymerization temperature (Tp) observed for the epoxy resin blended with benzoxazine in the absence of curing agent (hardener) was found to be around at 100°C. For example, the value of Tp obtained for the blends of C-a/Ep of 50:50 wt% is 106 °C and C-m/Ep of 50:50 wt% is 95 °C.

Oxirane ring is highly reactive than oxazine ring, which can be explained using primary, secondary and tertiary amines. Amines are highly reactive with oxirane ring functionality than that of oxazine ring. The curing temperature of benzoxazine/epoxy blends is lower than that of homo polymerisation. The oxirane ring forms a less stable intermediate of zwitterion formation with tertiary amine than that of oxazine ring, this influences to open the ring of benzoxazine with zwitterion intermediate. Hence, the polymerization of
benzoxazine occurs at lower temperature due to the influence of hydroxyl group bonded with oxirane intermediate, and in turn also contributes to form three dimensional cross-linked network polymer structure (Scheme 4).

**Thermal Conductivity**

The thermal conductivity is a material intrinsic property and dependent on its porosity because of their (volume of air/void content). It is also known that the values of density and thermal conductivity of materials are inter-related. The neat epoxy matrix possesses a higher value of thermal conductivity than that of the bio-material reinforced epoxy composites. This may be explained due to the fact that the bio-mass have a significant volume of air entrapped within the molecular structure because of their porous nature. The values of thermal conductivity obtained for the fibres reinforced benzoxazine/epoxy composites are presented in Table 1 and Fig. 2.

From the results obtained, it was inferred that the values of thermal conductivity are decreased with increasing the weight percentage concentration of bio-benzoxazine/epoxy matrix in the bio-mass reinforced composites. This may be explained due to the fact that the lower thermal conductivity fibrous materials than that of resinous bio-benzoxazine/epoxy matrix (Table 1).

**Thermal Resistance**

The values of thermal resistance obtained for rice husk and saw dust reinforced biobenzoxazine/epoxy composites are presented in Fig. 3. The value of thermal resistance is critically dependent on thickness and thermal conductivity of the samples. Since, all the composite panels prepared have almost the same thickness and the value of thermal resistance is inversely proportional to the thermal conductivity. However, the value of thermal resistance observed for bio-mass reinforced composites is higher than that of neat epoxy matrix. The comparison of values of thermal resistance obtained for the reinforced composite panels are presented in Table 1 and Fig. 3. It is also observed that the value of thermal resistance is increased, when increasing the fraction of the reinforcing bio-mass. Among the bio-mass used rice husk possesses the lower value of thermal conductivity (Table 1).
Table 1
Thermal and mechanical behaviour of bio-waste reinforced bio-benzoxazine/epoxy composites.

| Sample                       | Thermal Insulation behaviour | Mechanical behaviour |   |
|------------------------------|------------------------------|----------------------|---|
|                              | Thermal conductivity (W/m.k) | Thermal resistance (M² k/w) | Tensile strength (MPa) | Tensile modulus (GPa) | Elongation (%) | Hardness (HD) |
| Neat materials               |                              |                      |                |                  |                  |              |
| Neat epoxy matrix            | 0.0781                       | 0.0717               | 45.03          | 2.5651           | 1                | 84            |
| Poly(50 wt% C-a/50% Ep)      | 0.0628                       | 0.0796               | 7.79           | 0.3709           | 25               | 52            |
| Poly(50 wt% C-m/50% Ep)      | 0.0601                       | 0.0812               | 9.17           | 0.6683           | 17               | 65            |
| Rice husk reinforced composites |                              |                      |                |                  |                  |              |
| RH + Ep 100%                 | 0.0694                       | 0.0774               | 10.12          | 2.6057           | 1                | 80            |
| RH + (C-a 50% / Ep 50%)      | 0.0621                       | 0.0989               | 6.92           | 0.4282           | 3                | 51            |
| RH + (C-m 50% / Ep 50%)      | 0.0598                       | 0.1004               | 7.46           | 0.4986           | 3                | 72            |
| Saw dust reinforced composites |                              |                      |                |                  |                  |              |
| SD + Ep 100%                 | 0.0945                       | 0.0635               | 8.12           | 1.7042           | 1                | 68            |
| SD + (C-a 50% / Ep 50%)      | 0.0843                       | 0.0831               | 6.20           | 0.4478           | 3                | 47            |
| SD + (C-m 50% / Ep 50%)      | 0.0809                       | 0.0844               | 7.07           | 0.5492           | 3                | 58            |

**Mechanical Behaviour**

The values of tensile strength, tensile modulus, and elongation at break (%) of rice husk and saw dust reinforced benzoxazine/epoxy composites are determined in order to predict their strength behaviour and the results obtained are presented in Table 1 and Fig. 4. The values of tensile strength observed for rice husk and saw dust reinforced epoxy composites are 10.12 MPa and 8.12 MPa, respectively. Rice husk /
poly(C-m/Ep) composites possesses the higher values of tensile strength than that of rice husk/poly(C-a/Ep) composites due to its inherent strength behaviour (Table 1). Also saw dust/poly(C-m/Ep) composites possesses the higher values of tensile strength than that of saw dust/poly(C-a/Ep) composites. The elongation value of rice husk reinforced poly(C-m/Ep) composites is higher than that of rice husk reinforced poly(C-a/Ep) composites. Saw dust reinforced poly(C-m/Ep) composites is higher than that of saw dust reinforced poly(C-a/Ep) composites.

The values of hardness of bio-mass (rice husk and saw dust) reinforced bio-benzoxazine/epoxy composites were determined using Shore D Hardness test (Bergström 2015) and the results obtained are presented in Table 1 and Fig. 5. The rice husk and saw dust fibres reinforced bio-benzoxazine/epoxy composites were tested at ten different points on their respective surfaces and their average values were calculated. The rice husk and saw dust reinforced epoxy composites exhibited the highest value of hardness of 80 HD and 68 HD respectively. The values of hardness obtained for rice husk fibre reinforced poly(C-a/Ep) and poly(C-m/Ep) composites are 51, 72 HD, respectively. Similarly, the values of hardness observed for saw dust reinforced poly(C-a/Ep) and poly(C-m/Ep) composites are 47, 58 HD, respectively. It was observed that the values of hardness of the bio-mass reinforced epoxy composites are higher than that of bio-mass reinforced bio-benzoxazine/epoxy composites. Among the hybrid composite samples developed, the composites sample poly(C-a/Ep) and poly(C-m/Ep) possesses certain flexible behaviour than that of epoxy composites due to the presence of long chain alkyl moiety in cardanol.

**Acoustic Properties**

The acoustic properties of epoxy matrix and bio-mass (rice husk and saw dust) reinforced bio-benzoxazine/epoxy composites are determined using impedance tube setup in the region of low (50-1600 Hz) and high (500–6400 Hz) frequency using the circular specimens of 100mm and 29 mm having a thickness of ~ 5 mm and the results obtained are presented in Fig. 9. The composite samples reinforced with bio-materials having varied weight percentages of blend of bio-benzoxazine/epoxy matrices were developed and their sound absorption co-efficient (SAC) (Sabine 1929) was assessed in order to utilize them for acoustic proof panels. The values of SAC obtained for natural fibers reinforced biobenzoxazine/epoxy composites are increased up to 50-1600 Hz with 100mm and 500–6400 Hz with 29mm specimens of circular types.

Figures 6 and 7 present the acoustical performances of composites materials. The sound absorption behaviour of neat epoxy matrix observed over the range of frequency of 50-6400 Hz, whereas in the case of rice husk and saw dust reinforced epoxy composite samples show the higher values of absorption coefficient than that of neat epoxy matrix. The sound absorption behaviour is related to the internal structure of bio-mass. The presence of hollow tubular structure with numerous tiny pores contributes to reduce the transmission of vibrations by mechanical distribution through-out the material. Among the samples studied, the composite materials, rice husk and saw dust reinforced poly(C-m/Ep) composites possess the higher values of sound absorption coefficient than that of neat polymer matrix. The highest value of sound absorption coefficient of 6400 Hz (Fig. 6) was noticed in the case specimens developed...
using both bio-mass of rice husk and saw dust reinforced with hybridized blend of biobenzoxazine (50 wt%) and epoxy resin (50 wt%).

The test procedure described in the previous sections was used to predict the sound transmission properties of a biobenzoxazine/epoxy matrices and bio-mass reinforced blended biobenzoxazine/epoxy matrices composites. Transmission loss of sound (TSL) (Yahya 2009) is a measurement of the reduction in sound level of a sound source as it passes through an acoustic barrier. It is the number of decibels that are stopped by the acoustical barrier and is measured at different frequencies. Figure 7 shows the STL/frequency plots of neat and bio-mass reinforced biobenzoxazine/epoxy composite samples with dimensions of 29mm (higher frequency up to 6400 Hz) and 100 mm (lower frequency up to 1600 Hz) circular discs.

From Fig. 7a, it is evident that the values of sound transmission loss of lower frequency (1600 Hz) that observed for neat epoxy matrix, neat poly(C-a/Ep), neat poly(C-m/Ep), rice husk + Ep, rice husk + poly(C-a/Ep), rice husk + poly(C-m/Ep), saw dust + Ep and saw dust + (C-a/Ep), saw dust + poly(C-m/Ep), were 22 dB, 20 dB, 21 dB, 16 dB, 17 dB, 19 dB, 19 dB, 19 dB, 20 dB, 20 dB respectively. From Fig. 7b, it is also evident that the values of sound transmission loss of higher frequency (6400 Hz) that obtained for neat epoxy matrix, neat C-a/Ep, rice husk + Ep, rice husk + poly(C-a/Ep), rice husk + poly(C-m/Ep), saw dust + Ep, saw dust + poly(C-a/Ep) and saw dust + poly(C-m/Ep) were 31 dB, 32 dB, 16 dB, 18 dB, 20 dB, 22 dB, 21 dB and 30 dB respectively. Data obtained from sound transmission loss studies infer that these biobenzoxazine/epoxy blended samples can be utilized in the form of panels for acoustic insulation applications.

**Conclusion**

The importance of the present work is considered as the most of the materials used are either from sustainable bio-source or commercially available cost competitive materials. Cardanol-aniline (C-a) and cardanol-melamine (C-m) based benzoazinines were synthesised and blended with 50 wt% percentage of epoxy resin. The bio-benzoxazine/epoxy blended matrices prepared were reinforced with bio-mass (rice husk and saw dust) to obtain corresponding bio-composites and their thermal, mechanical and sound absorption properties were studied by appropriate techniques. Data obtained from different experimental results infer that the poly(C-m/Ep) based composites reinforced using rice husk and saw-dust possess an improved acoustic insulation performance than that of other samples. Data obtained from acoustic studies, it was observed that the highest value of sound absorption coefficient of 6400 Hz was noticed for composite specimens developed using both rice husk and saw dust reinforced with hybridized blend of biobenzoxazine and epoxy resin and these hybrid composite panels can be used as sound absorption material in the ceiling and wall construction applications.

**Declarations**

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Conflicts of interest/Competing interests

The authors have no conflicts of interest to declare that are relevant to the content of this article.

Availability of data and material

All the necessary data are included in the manuscript at appropriate section.

Code availability

Not applicable

Authors' contributions

Hariharan Arumugam: Conceptualization, Methodology, Investigation, Visualization, Data curation, Validation, Writing - original draft, Writing - review & editing.

Balaji Krishnasamy: Methodology.

A. Anto Dilip: Methodology.

M.I. Abdul Aleem: Resources, Validation, Supervision.

Muthukaruppan Alagar: Resources, Validation, Supervision.

Ethics approval

Not Applicable

Consent to participate

Not Applicable

Consent for publication

Not Applicable

References

Arumugam H, Krishnan S, Chavali M, Muthukaruppan A (2018) Cardanol based benzoxazine blends and bio-silica reinforced composites: Thermal and dielectric properties. New J Chem 42:4067–4080.
Asdrubali F, Ferracuti B, Lombardi L, et al (2017) A review of structural, thermo-physical, acoustical, and environmental properties of wooden materials for building applications. Build Environ 114:307–332. https://doi.org/10.1016/j.buildenv.2016.12.033

Balčiūnas G, Žvironaitė J, Vėjelis S, et al (2016) Ecological, thermal and acoustical insulating composite from hemp shives and sapropel binder. Ind Crops Prod 91:286–294. https://doi.org/10.1016/j.indcrop.2016.06.034

Bansod PV, Mittal T, Mohanty AR (2016) Study on the Acoustical Properties of Natural Jute Material by Theoretical and Experimental Methods for Building Acoustics Applications. Acoust Aust 44:457–472. https://doi.org/10.1007/s40857-016-0073-4

Belakroum R, Gherfi A, Bouchema K, et al (2017) Hygric buffer and acoustic absorption of new building insulation materials based on date palm fibers. J Build Eng 12:132–139. https://doi.org/10.1016/j.jobe.2017.05.011

Bergström J (2015) Experimental Characterization Techniques

Bhindare NH, Prakash S, Jatti VS (2019) A review on natural and waste material composite as acoustic material. Polym Test 106142. https://doi.org/10.1016/j.polymertesting.2019.106142

Binici H, Aksogan O, Demirhan C (2016) Mechanical, thermal and acoustical characterizations of an insulation composite made of bio-based materials. Sustain Cities Soc 20:17–26. https://doi.org/10.1016/j.scs.2015.09.004

Chen Y, Li D, Xu D (2019) Research on Application of Acoustic Materials in Automobile Noise Reduction. IOP Conf Ser Earth Environ Sci 233:. https://doi.org/10.1088/1755-1315/233/5/052026

Chourmouziadou K, Kang J (2008) Acoustic evolution of ancient Greek and Roman theatres. Appl Acoust 69:514–529. https://doi.org/10.1016/j.apacoust.2006.12.009

Elkhateeb AA (2012) The acoustical design of the new lecture auditorium, Faculty of Law, Ain Shams University. Ain Shams Eng J 3:219–235. https://doi.org/10.1016/j.asej.2012.04.005

Froimowicz P, R. Arza C, Han L, Ishida H (2016) Smart, Sustainable, and Ecofriendly Chemical Design of Fully Bio-Based Thermally Stable Thermosets Based on Benzoxazine Chemistry. ChemSusChem 1921–1928. https://doi.org/10.1002/cssc.201600577

Guna V, Ilangovan M, Hu C, et al (2019) Valorization of sugarcane bagasse by developing completely biodegradable composites for industrial applications. Ind Crops Prod 131:25–31. https://doi.org/10.1016/j.indcrop.2019.01.011
Hariharan A (2020) Studies on heterocyclic amines based cardanol-benzoxazine for oil-water separation. 1–13. https://doi.org/10.1002/pen.25396

Hariharan A, Kesava M, Alagar M, et al (2018) Optical, electrochemical, and thermal behavior of polybenzoxazine copolymers incorporated with tetraphenylimidazole and diphenylquinoline. Polym Adv Technol 29:355–363. https://doi.org/10.1002/pat.4122

Hariharan A, Prabunathan P, Kumaravel A, et al (2020) Bio-based polybenzoxazine composites for oil-water separation, sound absorption and corrosion resistance applications. Polym Test 86:106443. https://doi.org/10.1016/j.polymertesting.2020.106443

Hariharan A, Prabunathan P, Subramanian SS, et al (2019) Blends of Chalcone Benzoxazine and Bio-benzoxazines Coated Cotton Fabrics for Oil–Water Separation and Bio-silica Reinforced Nanocomposites for Low-k Applications. J Polym Environ. https://doi.org/10.1007/s10924-019-01629-2

Hariharan A, Srinivasan K, Murthy C, Alagar M (2017a) A Novel Imidazole-Core-Based Benzoxazine and Its Blends for High-Performance Applications. Ind Eng Chem Res 56:9347–9354. https://doi.org/10.1021/acs.iecr.7b01816

Hariharan A, Srinivasan K, Murthy C, Alagar M (2017b) Synthesis and characterization of a novel class of low temperature cure Benzoxazines. J Polym Res 25:. https://doi.org/10.1007/s10965-017-1423-0

He C, Huang J, Li S, et al (2018) Mechanically Resistant and Sustainable Cellulose-Based Composite Aerogels with Excellent Flame Retardant, Sound-Absorption, and Superantiwetting Ability for Advanced Engineering Materials. ACS Sustain Chem Eng 6:927–936. https://doi.org/10.1021/acssuschemeng.7b03281

Ismail AS, Jawaid M, Naveen J (2019) Void content, tensile, vibration and acoustic properties of kenaf/bamboo fiber reinforced epoxy hybrid composites. Materials (Basel) 12:. https://doi.org/10.3390/ma12132094

Kim HD, Ishida H (2001) Study on the chemical stability of benzoxazine-based phenolic resins in carboxylic acids. J Appl Polym Sci 79:1207–1219. https://doi.org/10.1002/1097-4628(20010214)79:7<1207::AID-APP80>3.0.CO;2-3

Kotzebue LRV, De Oliveira JR, Da Silva JB, et al (2018) Development of Fully Biobased High-Performance Bis-Benzoxazine under Environmentally Friendly Conditions. ACS Sustain Chem Eng. https://doi.org/10.1021/acssuschemeng.8b00340

Lakshmikandhan T, Hariharan A, Sethuraman K, Alagar M (2019) Development of functionalized SiO2–TiO2 reinforced cardanol and caprolactam modified diamine based polybenzoxazine nanocomposites for high performance applications. J Coatings Technol Res 16:1737–1749. https://doi.org/10.1007/s11998-019-00263-w
Liao J, Zhang S, Tang X (2020) Sound Absorption of Hemp Fibers (Cannabis Sativa L.) Based Nonwoven Fabrics and Composites: A Review. J Nat Fibers 00:1–13. https://doi.org/10.1080/15440478.2020.1764453

Liuzzi S, Rubino C, Martellotta F, et al (2020) Characterization of biomass-based materials for building applications: The case of straw and olive tree waste. Ind Crops Prod 147:112229. https://doi.org/10.1016/j.indcrop.2020.112229

Lyu Y, Ishida H (2019) Natural-sourced benzoxazine resins, homopolymers, blends and composites: A review of their synthesis, manufacturing and applications. Prog Polym Sci 99:101168. https://doi.org/10.1016/j.progpolymsci.2019.101168

Mamtaz H, Fouladi MH, Al-Atabi M, Namavivayam SN (2016) Acoustic absorption of natural fiber composites. J Eng (United Kingdom) 2016:. https://doi.org/10.1155/2016/5836107

Marques B, Tadeu A, António J, et al (2020) Mechanical, thermal and acoustic behaviour of polymer-based composite materials produced with rice husk and expanded cork by-products. Constr Build Mater 239:117851. https://doi.org/10.1016/j.conbuildmat.2019.117851

Mohammed F, Shaker D, Jalal N, et al (2018) Anbar Journal Of Engineering Science © Effect of Using Palm Wastes as Additive to Unsaturated Polyester on the Thermal and Acoustic Isolation Properties of a Composite Materials. 7:202–209

Muthukaruppan A, Arumugam H, Krishnan S, et al (2018) A low cure thermo active polymerization of chalcone based benzoxazine and cross linkable olefin blends. J Polym Res 25:. https://doi.org/10.1007/s10965-018-1556-9

Paun F, Gasser S, Leylekian L (2003) Design of materials for noise reduction in aircraft engines. Aerosp Sci Technol 7:63–72. https://doi.org/10.1016/S1270-9638(02)00006-8

Ručigaj A, Alič B, Krajnc M, Šebenik U (2015) Curing of bisphenol A-aniline based benzoxazine using phenolic, amino and mercapto accelerators. Express Polym Lett 9:647–657. https://doi.org/10.3144/expresspolymlett.2015.60

Sabine PE (1929) The measurement of sound absorption coefficients. J Franklin Inst 207:341–368. https://doi.org/10.1016/S0016-0032(29)91450-2

Sair S, Mansouri S, Tanane O, et al (2019) Alfa fiber-polyurethane composite as a thermal and acoustic insulation material for building applications. SN Appl Sci 1:1–13. https://doi.org/10.1007/s42452-019-0685-z

Sari NH, Wardana ING, Irawan YS, Siswanto E (2017) Corn husk fiber-polyester composites as sound absorber: Nonacoustical and acoustical properties. Adv Acoust Vib 2017:. https://doi.org/10.1155/2017/4319389
Selvaraj V, Jayanthi KP, Alagar M (2019) Livestock chicken feather fiber reinforced cardanol benzoxazine-epoxy composites for low dielectric and microbial corrosion resistant applications. Polym Compos 40:4142–4153. https://doi.org/10.1002/pc.25275

Shukla S, Yadav N, Lochab B (2017) Cardanol-Based Benzoxazines and Their Applications. Elsevier Inc.

Siano D, Viscardi M, Panza MA (2016) Automotive Materials: An Experimental Investigation of an Engine Bay Acoustic Performances. Energy Procedia 101:598–605. https://doi.org/10.1016/j.egypro.2016.11.076

Singh VK, Mukhopadhyay S (2020) Banana fibre-based structures for acoustic insulation and absorption. J Ind Text. https://doi.org/10.1177/1528083720901823

Taban E, Tajpour A, Faridan M, et al (2019) Acoustic Absorption Characterization and Prediction of Natural Coir Fibers. Acoust Aust 47:67–77. https://doi.org/10.1007/s40857-019-00151-8

Tang X, Yan X (2020) A review on the damping properties of fiber reinforced polymer composites. J Ind Text 49:693–721. https://doi.org/10.1177/1528083718795914

Tholkappiyana E, Saravanan D, Jagasthitha R, et al (2015) Modelling of sound absorption properties of sisal fibre reinforced paper pulp composites using regression model. Indian J Fibre Text Res 40:19–24

Thyavihalli Girijappa YG, Mavinkere Rangappa S, Parameswaranpillai J, Siengchin S (2019) Natural Fibers as Sustainable and Renewable Resource for Development of Eco-Friendly Composites: A Comprehensive Review. Front Mater 6:1–14. https://doi.org/10.3389/fmats.2019.00226

Trematerra A, Lombardi I (2017) Acoustic properties of cork sheets. Key Eng Mater 744 744 KE:66–70. https://doi.org/10.4028/www.scientific.net/KEM.744.66

Wang C, Sun J, Liu X, et al (2012) Synthesis and copolymerization of fully bio-based benzoxazines from guaiacol, furfurylamine and stearylamine. Green Chem 14:2799. https://doi.org/10.1039/c2gc35796h

Yahya I (2009) Analytical Expression for Sound Transmission Loss Calculation: An Improvement to the Existing Method after Singh and Katra. Adv Acoust Vib 2009:1–3. https://doi.org/10.1155/2009/574604

Zeng K, Huang J, Ren J, Ran Q (2019) Curing Reaction of Benzoxazine Under High Pressure and the Effect on Thermal Resistance of Polybenzoxazine. Macromol Chem Phys 220:1–9. https://doi.org/10.1002/macp.201800340

Zhan Z min, Yan H qiang, Yin P, et al (2020) Synthesis and properties of a novel bio-based benzoxazine resin with excellent low-temperature curing ability. Polym Int 69:355–362. https://doi.org/10.1002/pi.5957

Figures
Figure 1

Rice husk and saw dust reinforced benzoxazine/epoxy composites panel.

Figure 2

Thermal conductivity of (a) rice husk, (b) saw dust reinforced bio-benzoxazine/epoxy composites.

Figure 3

Thermal resistance of (a) rice husk, (b) saw dust reinforced bio-benzoxazine/epoxy composites.
Figure 4

Tensile strength and modulus of (a) neat bio-benzoxazine/epoxy matrix and (b) rice husk, (c) saw dust reinforced biobenzoxazine/epoxy composites.

Figure 5
Hardness of (a) Neat bio-benzoxazine/epoxy matrix, (b) rice husk, (c) saw dust reinforced bio-benzoxazine/epoxy composites.

Figure 6

Sound absorption coefficient of (a) Neat bio-benzoxazine/epoxy matrix, (b) rice husk, (c) saw dust reinforced bio-benzoxazine/epoxy composites.

Figure 7
Sound transmission loss of (a) lower frequency (b) higher frequency for rice husk and saw dust reinforced bio-benzoxazine/epoxy composites.

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