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Influence of Filler Size Variation on the Hardness, Thermal, and Moisture Absorption Properties of Wood Apple/Pine Cone Reinforced Areca Fiber Hybrid Composites

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Abstract

The ever-growing agricultural biomass has created a lot of burden on the already overburdened landfills, and has also led to increase in methane emissions from its burning. Therefore, the utilisation of this abundant biomass has become essential nowadays for the sustainable development of environment friendly materials. This work investigates the physicochemical and mechanical properties of lignocellulosic biomass reinforced polymer composite. In particular, a comparison was made between two sets of composites based on wood apple and pine cone fillers in terms of their size variation and its effect on their thermal, water absorption, and hardness properties. The results indicate that the filler size variation had a notable impact on the composite. Thermogravimetric analysis revealed that specific filler sizes improved thermal stability by limiting the polymeric chain mobility. In contrast, water absorption was found to increase with larger filler sizes due to particle agglomeration and void formation. The pine cone composites notably displayed higher moisture absorption than the wood apple composites due to the presence of higher hemicellulose content in pine cones. Contrarily, hardness values decreased as the filler size increased, attributed to agglomeration of fillers at large size. However, pine cone composites demonstrated superior hardness overall attributed to the higher amount of lignin present in pine cones. These findings highlight the necessity of optimizing filler size and selecting appropriate waste derivatives to balance thermal stability, moisture resistance, and mechanical strength in composites.

Keywords: Biomass, Waste utilisation, TGA, Water absorption, Hardness

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1 Introduction

The waste from landfills and other sources is responsible for 20% of methane emissions, which is the biggest contributor towards global warming, and is also a harmful air pollutant [1]. As predicted by the World Bank, the global waste generation is projected to rise by 70% as of 2050 unless suitable measures are adopted to curb it down [2]. The utilization of plastics and its based synthetic polymer can also be brought down by incorporating wastes as reinforcements, as this plastic waste is the major contributor towards the pollution of the water bodies and oceans that is hazardous to marine life and ecosystems [3]. A staggering 37% of waste generated throughout the world ends up in landfills, while a part of the remaining is being recycled, burned or open dumped [4]. The global waste management outlook 2024, published by UNEP, states that around 1000 chemicals utilised in plastic products processing and making are classified as problematic to endocrine system of the human body as well as carcinogenic [5]. The fashion and textile industries also generate an enormous waste and hence the United Nations is working on reducing this waste by emphasizing on advanced circular solutions for its profitable reuse [6]. In this 21st century, with the development of high-tech electronic products, the e-waste generation is on the rise. As per projected by UN for the year 2021, the average e-waste production by a single individual was estimated to be 7.6 kg. This type of waste is non-biodegradable and a part of it can be toxic in nature [7]. Another problem with utilizing plastics is the generation of microplastics generated from the breakdown of objects made up of plastics and synthetic textiles like bottles, packing materials, utensils, tyres etc. The biggest concern associated with microplastics is their entry into the water and soil and ultimately into our food chain [8–10]. To tackle these concerns industries and researchers are continuously striving for reducing the waste by using it in one from or other, or incorporating it as reinforcements in polymer composites, which not only reduces the weight fraction of polymer used in case of thermoset composites, but also reduces the cost associated with fabricating components made of composite materials. Many tech giants are also coming up with products that can lower their carbon footprint, like Apple unveiling its carbon neutral watch and Google utilising completely recycled aluminium for making the phone enclosures of their mobile phones namely Google Pixel 7 [11, 12]. The focus of manufacturing industries has also started to shift towards the usage of sustainable eco-friendly fibers/fillers reinforced polymer composites due to these reinforcements being lightweight, cost-effective, and abundance [13]. Wastes in the forms of reinforcements are now also being incorporated in natural fiber composites for improving their certain properties, like thermal stability at elevated temperatures, mechanical properties like tensile, flexural, and impact strength, and also their acoustic and properties. Owing to the growing utilisation of filler reinforced polymers in various applications replacing traditional or conventional parts like in packaging materials, biomedical parts, insulating materials etc., these reinforced polymers should possess good static and dynamic mechanical properties along with wear resistant. On one hand the lignocellulosic fibers and fillers have an inherent affinity for water molecules, the matrix is hydrophobic in nature and this negatively effects the interfacial adhesion between the reinforcements and the matrix resulting in ineffective stress transfer between the two [14, 15]. A good interaction between the reinforcement and the matrix is of prime importance for effective stress transfer and increased adhesion between the two. For this purpose, the fibers can be subjected to physical methods like plasma treatment that increase roughness of a fiber's surface thus improving adhesion or chemical methods wherein a fiber's is made more hydrophilic thus improving the adhesion [16].

Mahapatra et al. utilised waste textile fibers as reinforcements for fabricating epoxy-based composites for sound absorbing applications [17]. Human hair, pineapple leaf fiber, and areca fiber were utilised as reinforcements (15,20, and 25% vol. fraction) in the fabrication of fiber reinforced epoxy composites by Ali et al. [18]. For composites meant for outdoor applications, water absorption is a major issue that severely impact their service life. the lignocellulosic fillers and fibers are rich in cellulose and other constituents that have high affinity for water absorption [19]. The water absorption causes swelling in the composites, as well as severely decreases the interaction between the matrix and the reinforcements [20]. Despite the prior use of pine cones and wood apple shells in material science, their specific impact as varying-sized fillers in hybrid areca fiber composites represents a distinct gap in the literature. To address this, the current study leverages these abundant biomass sources alongside areca sheaths to determine how filler size dictates the thermal, water absorption, and hardness characteristics of the resulting reinforced composites.

2 Materials and Methods

2.1 Materials

Pine cones were collected from hills of Kumaon region, Uttarakhand, while wood apple shells were procured from local shop. Both the fillers were then grounded utilising a planetary ball mill, Retsch PM-400. The fillers were classified into different size range utilising a sieve shaker equipment in three different ranges of ASTM 200-ASTM 100 mesh, ASTM 100-ASTM 70 mesh, and ASTM 70-ASTM 50 mesh. For the reader's convenience, the fillers in the size range of 75-150 µm is referred as small, 150-212 µm as moderate, and 212-300 µm as large. Wood apple filler contains 39.54% cellulose, 26.06% hemicellulose, and 29.86% lignin, whereas pine cone comprises 18.8% cellulose, 46.5% hemicellulose, and 37.4% lignin [21, 22]. The resin and hardener utilised in this work were Lapox L-12 and Lapox AH-337 purchase from Atul Ltd. The resin to hardener mix ratio was 100:32 and the resin had a 28% bio-based carbon content as per the details provided by the supplier. The analytical measurement was cited as "percent modern carbon (pMC)." This is the percentage of C14 measured in the sample relative to a modern reference standard (NIST 4990C). The areca sheath fibers were procured from Go Green Products, Chennai, India. The images of both the fillers is shown in Figure 1.

Fig. 1: Images of fillers (a) Pine cone (left) (b) Wood apple (right)

2.2 Fabrication

Closed hand lay-up method was utilised for fabricating the composites. The weight percentage of areca sheath fibers was kept fixed at 30%, while fillers were added at 5 wt.%. The method of fabrication is explained in detail in our previously published work [23]. The specimens were cured at 70℃, followed by post curing for 24 hours. Composite designation is presented in Table 1.

Table 1: Composite's designation based on filler size

S. No.	Filler	Filler size (μm)	Composite Designation
1	Wood apple shell	75–150	A1
2	Wood apple shell	150–212	A2
3	Wood apple shell	212–300	A3
4	Pine cone	75–150	B1
5	Pine cone	150–212	B2
6	Pine cone	212–300	B3

2.3 Thermogravimetric analysis

The thermal degradation of composites was analysed using a PerkinElmer TGA apparatus. 10-15 milligrams of the sample was heated from room temperature to 600 ℃ at 30 ℃/min using a platinum pan placed under a nitrogen atmosphere having a gas flow rate of 20 ml/min.

2.4 Water absorption test

The absorption rate of composite specimens subjected to water absorption was evaluated as per ASTM D 570-98. The kinetics of water absorption of the specimens were carried out at room temperature, where the samples were weighed at regular intervals. Before weighing the specimens were wiped to remove the moisture present on the composite's surface. The percentage of water absorption (W) was calculated utilising equation 1.

$$W(\%) = \frac{W_2 - W_1}{W_1} \times 100\% \tag{1}$$

2.5 Hardness test

The hardness test of the composite specimens was carried out utilizing Milhard's Shore-D hardness equipment. The range (shore) of the equipment was from 0 to 100, and had a least count of 1. The average of five readings was reported for each type of specimen.

3 Results and discussions

3.1 Thermogravimetric analysis

Thermogravimetric analysis was performed for all the samples, in which weight loss was observed for all, although the rate of weight loss varied from sample to sample, depending upon the nature of reinforcement and size. The degradation of the samples can be visualized from Figure 2. Firstly, there was an initial loss of weight up to 150 ℃, which was due to the evaporation of moisture present in the samples. Further weight loss was due to the degradation of the constituents from which these reinforcements are consists of, namely cellulose, hemicellulose, and lignin. The increment of particle size had a positive impact on the thermal degradation behavior of wood apple-based composites initially, where the weight loss percentage of A3 composite was on the lower end in comparison to other composites. This could be due to a positive hybridization effect at that particular loading, where the wood apple fillers restricted the mobility of polymer chains at higher temperatures, which reduced its thermal degradation at higher temperatures to some extent. Zeng et al. [37] also reported an increase in thermal stability when loading different sized aluminium oxide fillers in epoxy matrix composites, also showing a positive hybridization effect on loading at larger sized filler loadings. The thermograms also revealed that the char yield was highest for A1 in case of wood apple composites and for B2 in case of pine cone composites. This indicated a higher thermal stability of both the composites at higher temperature. This higher thermal stability of both these composites at higher temperatures was also in accordance with their higher storage modulus and lower damping factor values as discussed in the previous sections. In the case of pine cone composites, as the particle size decreased from large to moderate, the weight loss percentage decreased. This could be due to the filler particles being properly dispersed in the matrix and preventing its degradation up to a certain extent more than the composites loaded with other sized pine cone particles.

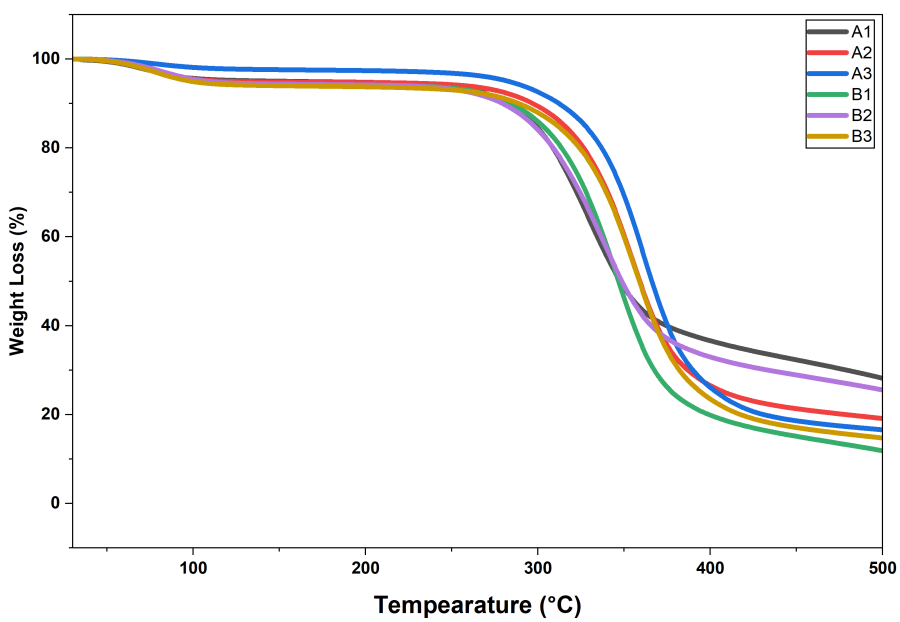

Fig. 2: TGA of the developed composites

3.2 Water absorption

Figure 3 shows the influence of reinforcement size variation on the water absorption characteristics of the hybrid composites. The water absorption in lignocellulosic reinforcement-based composites is due to the combination of several factors like cellulose/lignin content, fiber matrix interface, void content etc [24]. In the water absorption process of composites, the molecules of water get diffused in the specimen adhering to the hydrophilic components of lignocellulosic fillers and fibers by forming hydrogen bonds [25]. The water absorption was found to increase with the increasing filler sizes for both wood apple and pine cone reinforced composites. The water absorption behavior followed the same pattern for all the composites, initially being at a higher rate, and then getting stabilised, due to the composites getting saturated with water. The water absorption for composites reinforced with large sized wood apple and pine cone fillers was 10.19% and 11.76% higher than the composites reinforced with smaller sized fillers respectively. The increase in water absorption with increasing filler size could be due to an increased agglomeration and the resulting void spaces, which permitted more amount of water to penetrate [26, 27]. A similar pattern of increased water absorption with increasing filler size was reported by Bouafif et al. on incorporating wood particles of varying size in the composites at lower weight percentages, and was associated with the presence of a larger number of hydrophilic surfaces in larger sized particles [28]. On comparing the water absorption behavior between the pine cone and wood apple-based composites, it was found that pine cone composites absorbed a higher amount of water. this was due to the presence of comparatively large amount of hemicellulose in the pine cone particles in comparison to the wood apple particles. Hemicellulose due to its amorphous structure have hydroxyl groups exposed and accessible to the water molecules resulting in the increased water absorption [29].

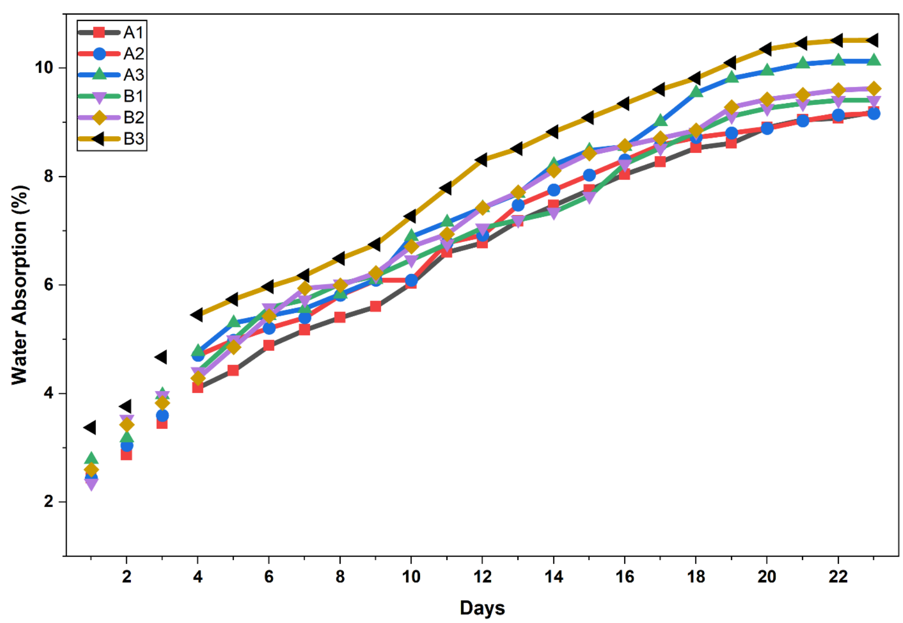

Fig. 3: Water uptake behavior of developed composites

3.3 Hardness

The hardness (Shore-D) of a natural fiber-based composite is dependent on the lignocellulosic filler's chemical composition, size, and weight percentage. As evident from Figure 4, hardness of the composites was greatly influenced by the size variation of both the fillers. The hardness for both set of composites decreased as the size of the fillers increased. This increase in hardness value. The decrease in hardness value as the particle size was increased from small to large can be due to the filler agglomeration and ineffective stress transfer between the reinforcement and the matrix. These large sized fillers have a tendency to form clusters so as to reduce the surface energy interactions with the matrix, while the ineffective stress transfer can stem from lesser interfacial bonding between the fillers and the matrix [30, 31]. On comparing the hardness values of wood apple and pine cone based composites, it was found that the pine cone fillers had a higher hardness. This was due to the higher amount of lignin present in the pine cone fillers which resist the localized plastic deformation effectively. It also increases the compressive strength of the fillers by binding its cell structure tightly [32].

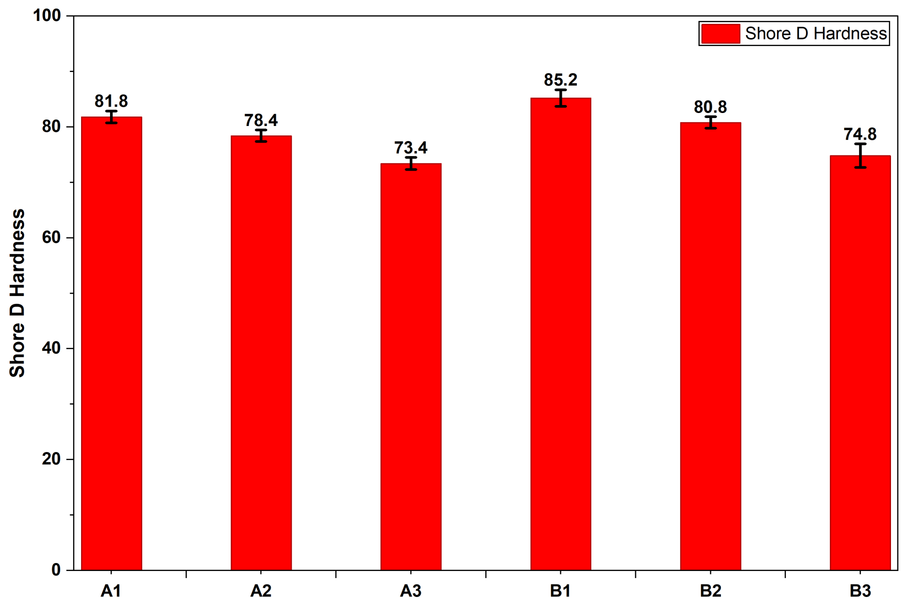

Fig. 4: Shore-D hardness of developed composites

4 Conclusion

In the present study, experimental investigation on the influence of filler size on the thermal, absorption, and hardness properties was carried out. All the reinforcements utilised in this work forms a part of agricultural biomass and an effort was made to utilise them in fabrication of composites. Apart from this, a comparison between two different lignocellulosic fillers-based composites was made, and their thermal, hardness, and water absorption properties was carried out. Both the fillers (wood apple and pine cone) were sieved to different size range with the help of sieve shaking apparatus. Incorporating different sized in the composites had a notable impact on the different properties of the developed composites. The thermal stability of the composites was significantly affected by the filler type and its size as revealed by thermogravimetric analysis. The thermal degradation followed the usual procedure, initial moisture evaporation up to 150 ℃ followed by the breakdown of the lignocellulosic contents (cellulose, hemicellulose, and lignin). The increase in filler size improved the thermal resistance suggesting that larger fillers restricted effectively the mobility of polymeric chain at higher temperatures. The higher char yield was found for

A1 and B2 composites. The water absorption behavior of the developed composites was also governed by the filler size and chemical composition. There was an increase in water absorption associated with larger filler size that was attributed to the agglomeration of fillers and voids. Pine cone composites were found to be more susceptible to water uptake because of higher hemicellulose content. The hardness on the other hand decreased as the filler size increased for both set of composites. Better dispersion facilitated effective stress transfer between the reinforcement and the matrix. Furthermore, a high lignin content resulted in the pine cone fillers-based composites having more hardness than the wood apple composites. Future investigations should prioritize chemical surface modifications (e.g., alkalization or silane treatments) of these highly lignified particulates to mitigate the interfacial debonding and fiber pull-out observed in the fractographic analysis.

Declarations

- The authors received no specific funding for this study.
- The authors declare that they have no conflicts of interest to report regarding the present study.
- No Human subject or animals are involved in the research.
- All authors have mutually consented to participate.
- All the authors have consented the Journal to publish this paper.
- Authors declare that all the data being used in the design and production cum layout of the manuscript is declared in the manuscript.

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