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Enhancing mechanical properties of waste expanded polystyrene composites through varied coupling agents and wood powder formulations

CahyoBudiyantoro^{1*}, FerriawanYudhanto²

¹Mechanical Engineering Department, Universitas Muhammadiyah Yogyakarta, Yogyakarta, 55183, Indonesia

²Automotive Engineering Technology Department, Universitas Muhammadiyah Yogyakarta, Yogyakarta, 55183, Indonesia

*Corresponding author: cahyo_budi@umy.ac.id

Abstract

This study investigates Wood Plastic Composites (WPCs) by incorporating waste Expanded Polystyrene (EPS) and various wood powder reinforcements. The mechanical properties of WPCs play a pivotal role in sustainable material development. Our research delves into the effects of different treatments on wood powders and their interactions with the polymer matrix. Pine, teak, and silk tree wood powders underwent alkali immersion and coupling agent treatments. The ensuing composites underwent rigorous testing, including flexural strength, hardness, and impact resistance assessments. The findings underline the complex factors governing WPC mechanical properties. Pine-based WPCs, reinforced with pine wood powder and subjected to alkali treatment, exhibited the highest flexural strength at 29.56 MPa, whereas the lowest flexural strength of 14.65 MPa was observed in WPCs reinforced with alkali-treated teak wood powder. The highest impact strength quantified at 2.54 kJ/cm², was found in untreated pine wood powder-based WPC. In contrast, the lowest impact strength was identified in teak wood powder-based WPC treated with alkali.

Keywords:

Wood plastic composite, expanded polystyrene, wood powder, mechanical properties.

1 Introduction

Expanded Polystyrene (EPS), also referred to as styrofoam, is a standard variety of Polystyrene (PS) that has been used in a wide range of applications, such as impact reduction wrapping, protective helmets, structural crushability, building products for filling in roadways, insulated concrete frameworks, and lightweight EPS foamed concrete. This material comprises nearly 98% air and 2% plastic [1]. Due to its low production cost and ease of manufacturing, EPS has been widely produced. However, the lifespan of this product is very short, and within a short period, it ends up in landfills. Being a non-degradable product, the large volume of EPS waste can harm the environment if not reused, recycled, or upgraded after use. The significance of EPS recycling contrasts with the numerous challenges it presents. EPS has a large volume coupled with low density (10–25 kg/m³), which creates difficulties in recycling [2].

Similar issues also occur with waste generated by the wood industry. Sawmill waste is not optimally utilized and is often discarded or burned. Indonesia's total sawn timber production in

2020 reached 2,580,000 m³ per year [3]. If sawdust accounts for approximately 10.6% of the total production [4], it produces approximately 258,000 m³ of sawdust waste annually.

Both materials can be formed into an alternative material by utilizing EPS as the matrix and wood powder as the filler in an innovative composite material called Wood Plastic Composite (WPC). WPC, a novel eco-friendly material, employs cellulose fibers as reinforcement agents within composite materials relying on polymer matrices and has found extensive application across diverse sectors, including architecture and parquet flooring [5]. Wood is a blend of polymers comprising partially crystalline cellulose microfibrils alongside sizable amorphous hemicellulose and lignin molecules [6]. However, there are several limitations found in WPC. Wood content makes it more hygroscopic and poses a higher risk of biological degradation [7][8]. Furthermore, compared to natural wood, the production cost of WPC is higher [9]. The properties of WPCs depend on the wood flour content, coupling agent, wood species, and plastic matrix.

Common thermoplastics for WPC manufacture are Polypropylene (PP), Polyethylene (PE), and Polyvinyl Chloride (PVC) [10][11]. Polystyrene, especially in EPS, is not widely used as a matrix in WPC. EPS cannot be directly used as a matrix; it must be converted into a solid state through a densification process involving thermal condensation and extrusion, reducing approximately 1/90 of the foam volume [12].

The main disadvantage of WPC is the low compatibility between the hydrophilic wood filler and hydrophobic polymer matrix. The interfacial bonding between the two components is crucial for transferring stress from the matrix to the reinforcement, and this bond can be enhanced by surface treatment; one of the surface treatment methods is the use of coupling agents on the polymer matrix [13]. The coupling agent produces functional groups inside the polymer chains of the matrix and then builds chemical or physical interactions with the reinforcing filler. Maleic anhydride is one of the most efficient modification agents to functionalize styrene polymers. Styrene Maleic Anhydride (SMA) is a famous coupling agent widely used in WPC manufacturing [14][15]. Coupling agents serve as hydrophobic media that provide stability and uniform dispersion of particles. The viscosity of the fluid decreases with the presence of coupling agents [10].

The type of wood powder also plays a role in compatibility, effective interaction area, and mechanical properties. Hardwood contributes to increased tensile strength and thermal deflection resistance compared to softwood [16]. Wood types with higher interfacial area enhance mechanical interlocking between the polymer and wood powder [17][18]. The microstructure of the wood type influences the increase or decrease in the interaction area. The polymer's molecular weight also influences the polymer's penetration level into the wood structure. Components with lower molecular weight can more easily penetrate the wood structure but may affect the decrease in mechanical properties of the composite. The wood powder's hygroscopic nature also affects WPC's performance [19][20]. In addition to enhancing the mechanical strength of WPC, coupling agents also contribute to increased moisture resistance [21][22].

The wood powder's shape and size impact WPC's flow characteristics and mechanical properties [23]. Sawdust cannot be used directly and requires smoothing and filtering steps to achieve the desired particle size. The particle size used in WPC typically varies from 40 to 80 mesh [24][25] when investigating the influence of particle size on the mechanical properties of wood/PP composites.

The production of WPC involves several stages, including material preparation, pellet manufacturing, dog bone specimen manufacturing, mechanical testing, and morphological observation. Material preparation includes the preparation of EPS matrix and wood powder. EPS waste needs to be transformed into

solid PS particles through mechanical crushing, followed by melt densifying to convert foam powder into solid PS blocks, which are then crushed into solid PS particles. Sawdust is filtered to achieve the desired mesh size, soaked in NaOH, and dried [26].

Pellets are created by mixing wood powder, plastic, and a coupling agent at the initial step of manufacture. Injection or hot press molding is then used for processing the pellets to produce specimens or products [27].

The primary goal of this study was to contribute to the field of WPC materials by addressing two significant challenges. The first challenge is utilizing waste EPS as the primary matrix, unlike traditional WPCs that typically use common polymers like PP, PE, or PVC as the matrix, this research focused on repurposing waste EPS, which is often underutilized and ends up in landfills. We intended to demonstrate that EPS, despite its low density and non-standard use as a matrix, can effectively serve as the matrix in WPCs. This approach has the potential to not only reduce EPS waste but also provide a more sustainable solution for composite materials. Secondly, investigating treatment variations and wood powder types to expand the understanding of how treatment processes (such as styrene maleic anhydride, bleaching, and no treatment) and the choice of wood powder types (teak, pine, and silk tree) impact the mechanical properties of WPCs. This research provides valuable insights into optimizing treatment methods and wood powder selection to enhance the performance of wood-based composites. This study investigates the influence of treatment variations and types of wood powder on the mechanical properties of wood powder-reinforced waste-expanded polystyrene composites. Three treatments were applied: styrene maleic anhydride, bleaching, and no treatment. The wood powder was obtained from three commonly found wood types: teak, pine, and silk tree (*Albizia chinensis*). The WPC specimens were tested for bending strength, tensile strength, and impact resistance. The morphology of the specimen cross-sections was observed using a scanning electron microscope.

2 Materials and Methods

2.1 Materials

The EPS waste was collected from collectors at the final disposal location; it had a Melt Flow Index (MFI) of 11 g/10 min [2]. Waste is cleansed, dried, and then shredded to reduce its size. The wood powder used was derived from teak, pine, and silk tree wood and is sieved to have particle sizes ranging from 50 to 150 micrometers. Table 1 shows a comparison of the properties of three types of wood powders used in this study [28][29][30].

Table 1. Properties of wood

Properties	Unit	Wood type		
		Pine	Teak	Sengon
Moisture content	%	12.6	16.8	13.13
Average Density	gr/cm ³	0.58	0.67	0.56
Hardness	Kg/cm ²	175.5 – 285.8	414 - 428	286.91
MOE	Kg/cm ²	43998	127700	100499
MOR	Kg/cm ²	461.05	308.68 – 558.3	587.56

There are three types of treatments performed on the wood powder and matrix. The initial treatment is Styrene Maleic Anhydride (SMA) from Merck Company, Darmstadt, Germany [31]. This coupling agent, Aldrich S458066, comprises 30% maleic anhydride groups and has an average molecular weight of 7500 g/mol. The second procedure involves the application of bleaching to the wood powder, whereas the third variety does not include any treatment.

2.2 Specimen Preparation

Fig. 1 outlines the process of preparing a WPC specimen. The EPS waste underwent size reduction, with dimensions ranging from 2–5 cm, followed by heating at 120°C for 15 minutes. This heating step induced a 95% reduction in foam dimensions by initiating the softening of EPS, a phenomenon activated within the temperature range of 100°C to 120°C. The compacted EPS pieces were then shredded into smaller sizes to facilitate better mixing with the wood powder and processed to create specimens. The size of the wood particles is standardized within the range of 40–50 mesh through a sieving process.

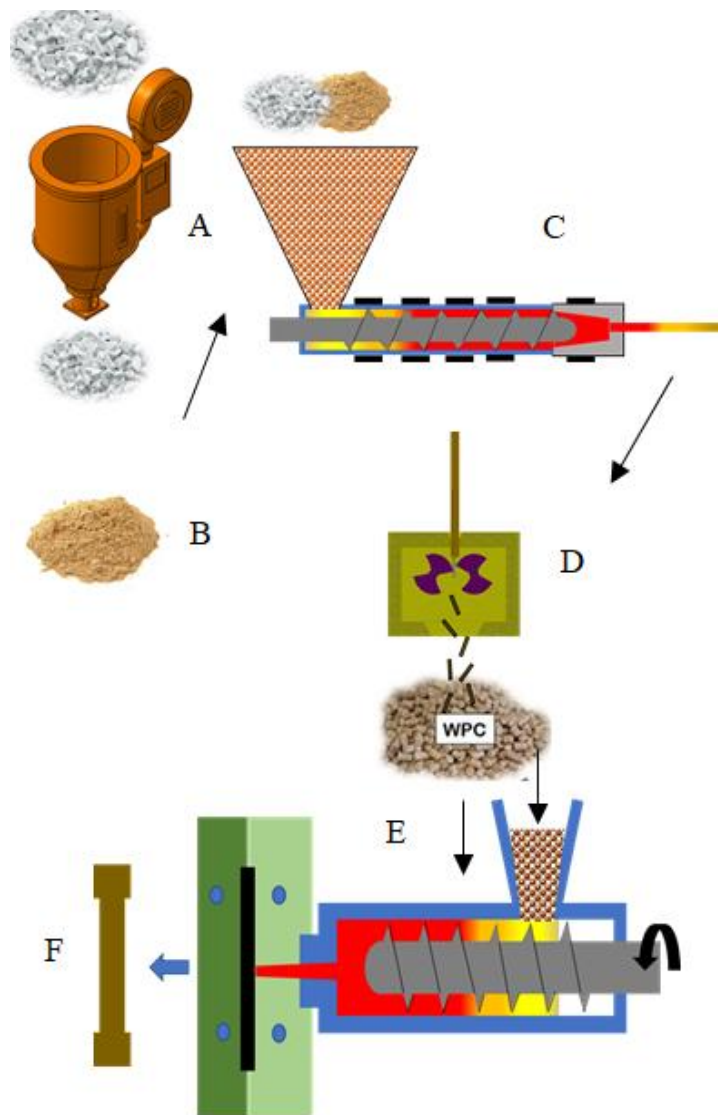


Fig. 1. WPC manufacturing steps.

Where:

- A : EPS densifying
- B : Wood flour treatment
- C : Wood – plastic compounding
- D : WPC filament cut into pellet
- E : Injection molding
- F : WPC dogbone specimen

Table 2 illustrates the experimental design with variations in treatments and types of wood powder, encompassing nine experimental runs. For experimental runs 1 to 3, WPC consists of 80wt% EPS and 20wt% wood powder. The bleaching process involved immersing the wood powder for 5 hours in a solution comprising 5% NaOH and 95% distilled water. Subsequently, the wood powder was dried at 160°C for 7-8 hours until completely dry [32]. In experiments 4 to 6, the wood powder underwent the

bleaching treatment, while the EPS received the coupling agent treatment. The EPS was immersed in a solution containing 2% SMA and 98% acetic acid. After the immersion, both materials were dried. The content of 2wt% SMA has been proven to provide satisfactory interfacial adhesion between wood flour and the polymer matrix [33]. After the immersion, both materials were dried. The EPS/wood flour composites were compounded using a screw extruder with an L/D ratio of 32. The barrel temperature was controlled between 130°C and 160°C [34] and the rotor speed was set at 40 rpm, respectively. From this process, plastic pellets containing wood powder are produced. The plastic pellets are processed in a 70 ton MEIKI injection molding machine to form tensile and impact test specimens at a barrel temperature of 160°C and a mold temperature of 40 ± 2°C, with a screw speed of 30 rpm and pressure of 850 bar.

Table 2. The research designs

Run	Wood powder	Treatment	Abbreviation
1	Pine	Untreated	WPC P-U
2	Pine	Alkalization	WPC P-A
3	Pine	Alkalization + SMA	WPC P-ACA
4	Teak	Untreated	WPC T-U
5	Teak	Alkalization	WPC T-A
6	Teak	Alkalization + SMA	WPC T-ACA
7	Sengon	Untreated	WPC S-U
8	Sengon	Alkalization	WPC S-A
9	Sengon	Alkalization + SMA	WPC S-ACA

2.3 Testing and Observation

Three mechanical property tests were conducted on the WPC specimens, namely the bending, impact, and hardness tests. For each variation, five repetitions were performed for each type of mechanical test. Static bending tests were conducted following ISO 178. A three-point bending strength test was conducted using a Zwick/Roell Z020 universal testing machine (manufactured in Germany) with a crosshead speed of 5 mm/min and a span length of 80 mm. The hardness test used the Shore D method according to ASTM D 2240. Impact testing refers to the Charpy method following ISO 179 standards with the flatwise specimen orientation. The mechanical properties of the experimentally produced WPC will be compared with specimens of compacted and injection-molded EPS and specimens made from PS material.

The morphological structure was observed using a Scanning Electron Microscope (SEM) model: Zeiss EVO 10. This analysis was used to study the filler–matrix adhesion as well as the fracture behavior of the composites. A section of the selected tensile fractured surface was coated with a thin palladium layer before analysis.

3 Results and Discussion

3.1 Molding Issues

During molding, the reduction in holocellulose content in wood fibers was observed to correlate with elevated temperature. This phenomenon can be attributed to the capacity of high temperatures to eliminate the acetyl group from the semi-cellulose, leading to the generation of acetic acid and more extensive degradation. In contrast, lignin experiences only minor

softening rather than degradation due to its excellent thermal stability [5]. For some reason, the residual content of lignin and the tendency of sticky waste Expanded Polystyrene (EPS) present challenges during the injection molding of Wood-Plastic Composite (WPC) samples. Lignin's stickiness can be explained from a chemical perspective by considering the types of chemical bonds and linkages present in its complex structure. Lignin is rich in phenolic compounds that can undergo various chemical reactions, including oxidation. The oxidative polymerization of phenolic compounds can lead to the formation of adhesive materials. Some phenolic compounds, like tannins, are naturally sticky and are used in adhesives. The residual lignin content can lead to a short shot during the molding of WPC specimens. The short shot happens because lignin, although a crucial component in wood, does not melt or flow easily during the molding process. When the WPC is melted and injected into the mold, the lignin particles can resist melting and flowing, creating areas of incomplete filling in the mold. These areas with insufficient material result in a short shot, where the final product lacks full coverage or volume, leading to defects in the molded specimen. In essence, lignin can hinder the uniform flow of the molten composite material within the mold, causing incomplete filling and short shots during molding.

Moreover, EPS can absorb moisture from the environment, leading to steam generation and expansion when subjected to high temperatures during injection molding. This can cause the material to become sticky, affecting its flow and causing defects in the final product. The sticky nature of degraded lignin and waste EPS can cause the WPC melt to adhere to the surfaces of the injection mold, resulting in difficulties during demolding.

Those two conditions can disrupt the flow of molten plastic into the mold. This can create physical barriers and hinder smooth flow throughout the mold. As a result, the molten plastic may not be able to adequately fill all areas of the mold (a phenomenon known as short shot), as illustrated in Fig. 2.



Fig. 2. Short shot phenomenon.

Alkalization treatment, such as immersion in alkaline solutions like NaOH, can alter the surface properties of wood powder. This treatment can reduce lignin content and enhance the surface affinity of the wood powder towards the polymer matrix, improving the adhesion between the two materials during the molding process.

The specimens were produced according to the research design in Table 1. Additionally, specimens without wood powder (consisting solely of injected EPS) were prepared as a comparison. Fig. 3 depicts the molded results of specimens with and without wood powder, with the former exhibiting a darker physical appearance.



(a)



(b)

Fig. 3. Molded specimens: (a) without wood powder, (b) with wood powder.

3.2 Mechanical Properties

3.2.1 Flexural Strength

The results of the flexural testing are presented in Fig. 4. It can be observed that the highest flexural strength value of 29.56 MPa was obtained for WPC reinforced with pine wood powder and subjected to alkali treatment. Alkali treatment can enhance flexural strength in WPCs with pine wood powder reinforcement. In this case, alkali treatment strengthens and stiffens the cellulose fiber content, thereby improving the adhesion between the composite-forming components [35]. However, when alkali treatment is combined with Styrene Maleic Anhydride (SMA), it might decrease flexural strength. In this case, the combination of alkali treatment and SMA leads to excessive rigidity that can reduce the composite's ability to withstand bending forces, resulting in reduced flexural strength. The different conditions observed in WPC with teak wood powder reinforcement indeed show a unique behavior. The initial alkali treatment causes a decrease in WPC strength to 14.65 MPa, which is even lower than specimens without reinforcement. However, when combined with Styrene Maleic Anhydride (SMA), the strength of the WPC improves to 25.13 MPa. Alkali treatment might have initially caused structural changes or degradation in the teak wood powder. Adding SMA could potentially stabilize or reinforce the altered teak wood structure. The flexural strength of WPC with teak wood powder and silk tree wood powder reinforcement is lower than that of WPC with pine wood powder reinforcement.

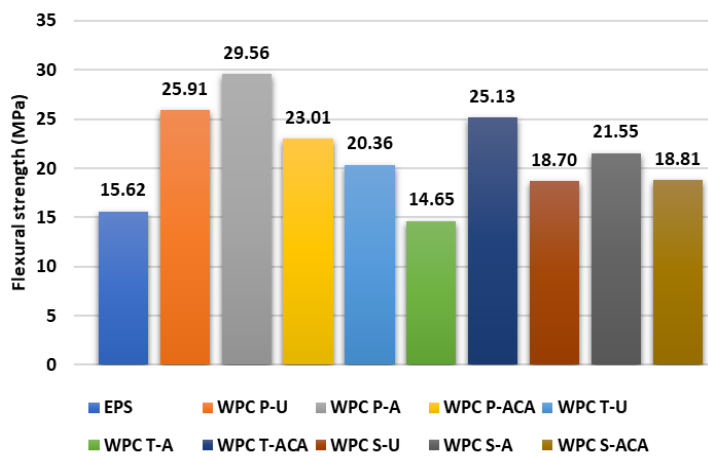
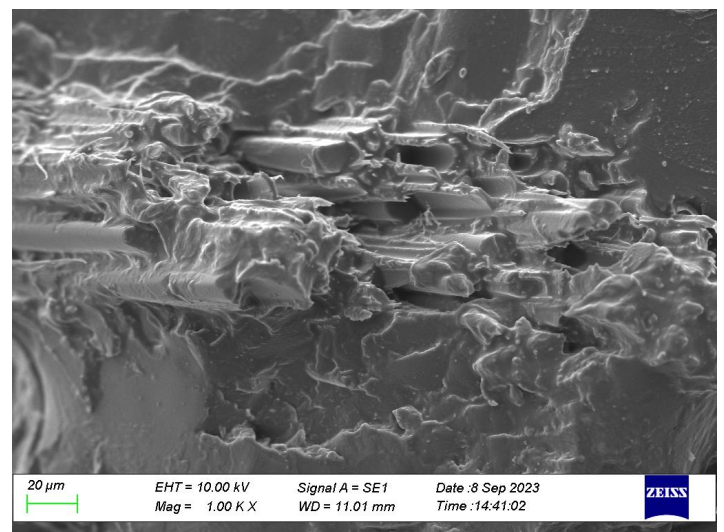


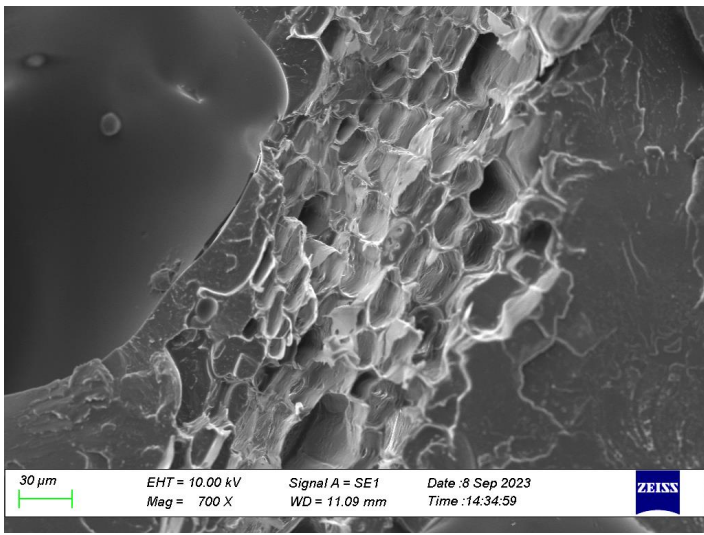
Fig. 4. Flexural strength of WPC.

The improvement in flexural strength of EPS-based Wood Plastic Composite (WPC) reinforced with pine wood powder can be attributed to several factors. One crucial factor is the wood powder and polymer matrix interaction. The interaction between pine wood powder and the polymer matrix (EPS) is likely more favorable compared to other types of wood powder. This can be due to better adhesion between pine wood powder and the polymer matrix, enhancing load transfer and resistance to flexural forces. This would need further confirmation through morphological observations on specimen cross-sections using a Scanning Electron Microscope (SEM), as can be seen in Fig. 5 and Fig. 6.

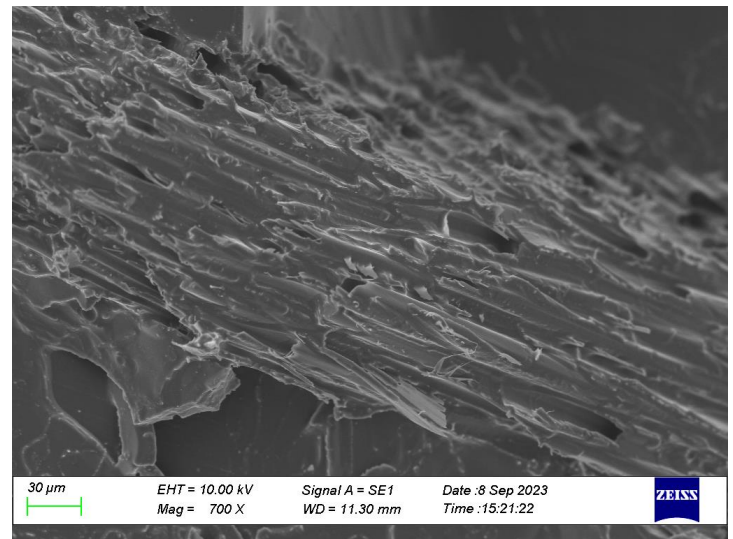
The flexural strength of WPC is highly dependent on the adhesive interaction between the reinforcing fibers and the matrix. Alkali treatment of pine fibers enhances the compatibility of the bond with the EPS matrix, resulting in the highest flexural strength for WPC P-A at 29.56 MPa. The matrix surrounding the wood powder demonstrates the adhesive capability, as seen in Fig. 5(a). Different conditions are observed in the wood powder reinforcement without treatment. The traces of cavities in Fig. 5(b) indicate that the wood powder easily detaches from the matrix when subjected to flexural loads on the specimen. Sequential treatment (alkalization and coupling agent) does not show any improvement in adhesion; the wood powder is also easily pulled out from the matrix (Fig. 5(c)).



(a)

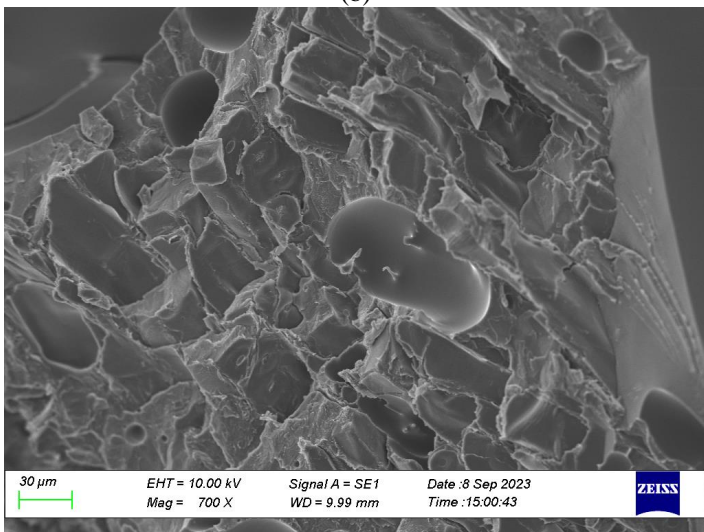


(b)



(b)

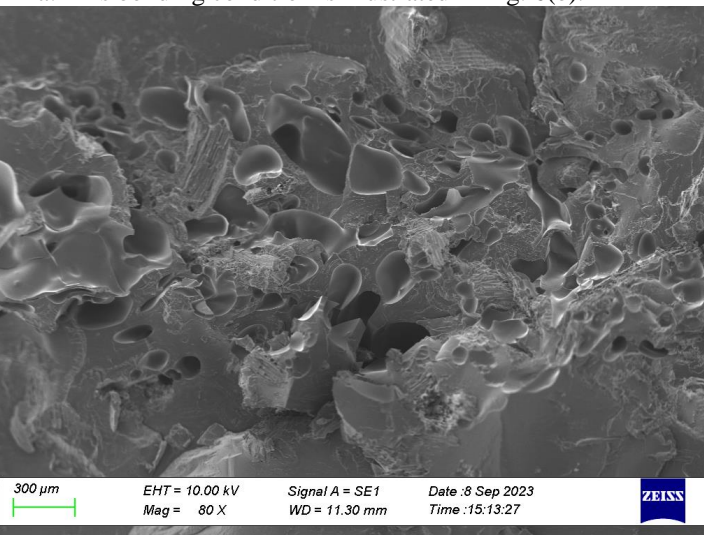
Fig. 6. SEM images of fractured surface: (a) WPC T-A, (b) WPC T-ACA.



(c)

Fig. 5. SEM images of fractured surface (flexural loaded): (a) WPC P-A, (b) WPC P-U, (c) WPC P-ACA.

Alkalized teak wood powder-reinforced WPC provides the lowest flexural strength, 14.65 MPa. Old teak wood still contains a significant amount of lignin even after the alkali treatment, where lignin cannot adhere well to the matrix. The fracture surface condition in Fig. 6(a) indicates numerous voids from the release of wood powder when subjected to flexural loading. Nevertheless, the sequential treatment (alkalization and coupling agent) can enhance the adhesion between wood powder and the matrix, thereby improving the flexural load-bearing capacity to 25.13 MPa. This bonding condition is illustrated in Fig. 6(b).



(a)



Fig. 7. Shear fracture due to flexural load.

3.2.2 Impact Strength

Impact strength testing using the Charpy method was conducted on a Zwick/Roell HIT 5.5P machine with a theoretical impact velocity of 2.901 m/s and a total mass of 0.2377 kg. The specimen was positioned flatwise about the pendulum strike direction. The impact strength, expressed in J/m² or kJ/m², provides information about the specimen's resistance to shock loads or how effectively it can withstand shocks without fracturing. The test results are presented in Fig. 8.

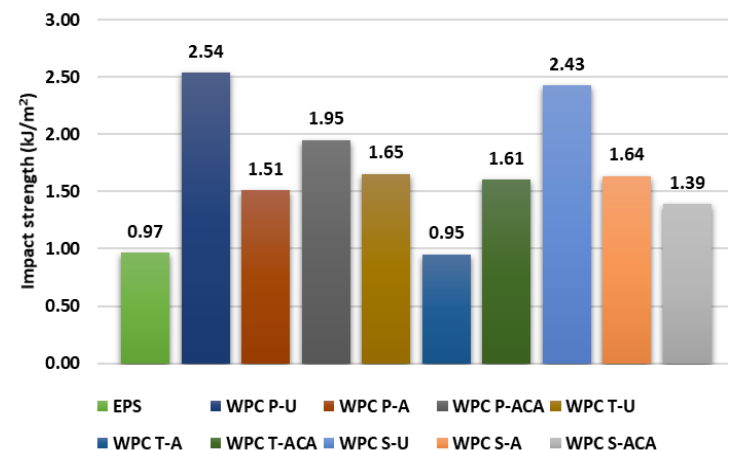


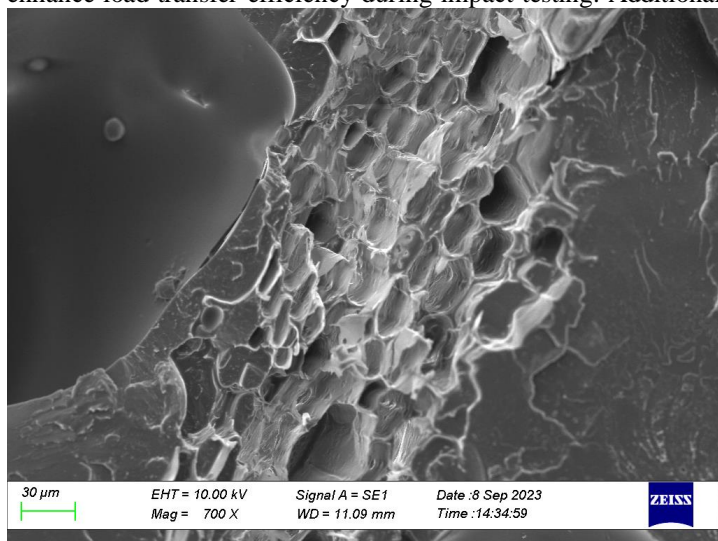
Fig. 8. Impact strength of WPC.

As depicted in Fig. 8, all treated composite specimens' impact strength was lower than untreated WPC. WPC reinforced with untreated pine wood powder (WPC P-U) displayed the highest impact strength, reaching 2.54 kJ/m², followed by WPC reinforced with untreated sengon wood powder (WPC S-U) at 2.43 kJ/m². This can be attributed to the wood particles functioning as stress concentrators, decreasing impact strength [36]. The residual lignin content on the untreated wood powder is likely contributing to WPC's toughness and impact resistance. Lignin can help maintain the structural integrity of wood powder and the polymer matrix when facing impact forces. When lignin is lost through treatment processes or thermal degradation, wood-plastic composites tend to lose some of their mechanical properties associated with lignin. This can lead to a decrease in impact strength, as the impact resistance and toughness provided by lignin are no longer present to the same extent. In the case of the teak wood powder reinforcement, alkali treatment is observed to decrease the impact strength of the WPC due to the reduction in lignin content. Compatibility between alkali-treated teak wood powder and the EPS polymer matrix also plays a crucial role in material interaction [37][38]. Sequential treatment involving alkali and Styrene Maleic Anhydride (SMA) can enhance compatibility between teak wood powder and the polymer matrix. SMA is a coupling agent that can improve the adhesion between the wood powder and EPS matrix. This combination of treatments may enhance load transfer efficiency during impact testing. Additional

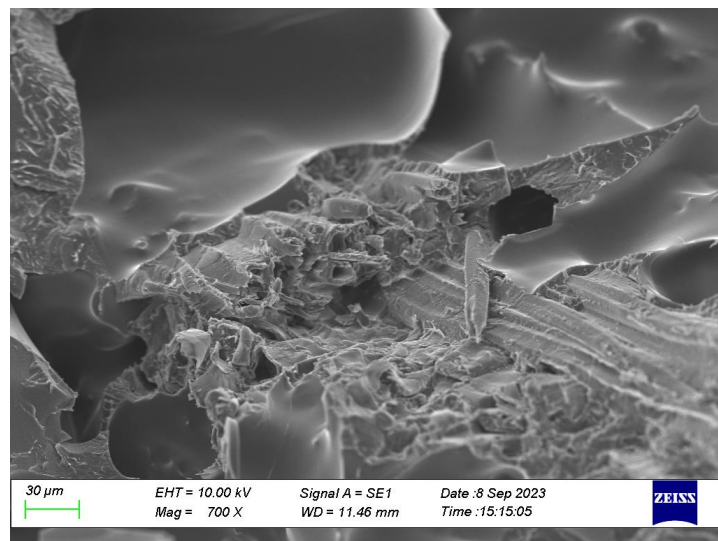
research is needed to investigate the effect of fiber loading and lignin content on impact strength.

Fig. 9 displays SEM images of the impact-fractured surface of the WPC specimen. Fig. 9(a) shows the fracture condition due to impact loading for WPC specimens with untreated pine wood powder reinforcement. Resistance to impact loads, in this case, is dominated by the interaction of the remaining lignin content with the matrix rather than the adhesive interaction of the wood powder. Some portions of the wood powder experience pull-out during impact. Furthermore, in Fig. 7(b), many formations of voids of various sizes are found on the WPC-reinforced alkaliized teak wood powder. Due to their incompatibility, such morphologies might have resulted from a weak interfacial adhesion between the EPS matrix and teak wood powder during alkalinization. This condition leads to a decrease in impact resistance.

The fracture surface caused by the impact load on WPC reinforced with sengon wood powder is depicted in Fig. 10. There does not appear to be a strong bond between untreated wood powder and the matrix in Fig. 10(a). Impact strength is dominated by the distribution of wood powder and the role of the matrix. The bonding conditions improve when the wood powder is treated with an alkali and a coupling agent, as seen in Fig. 10(b). However, the bond easily detaches and, therefore, does not improve impact resistance.

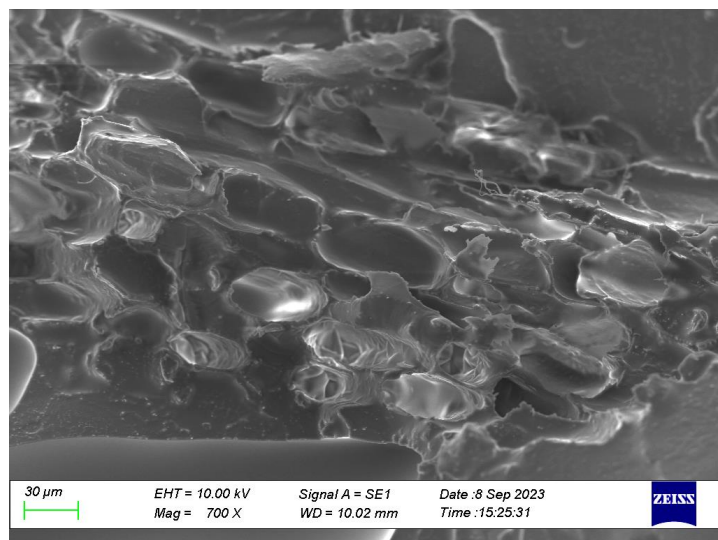


(a)

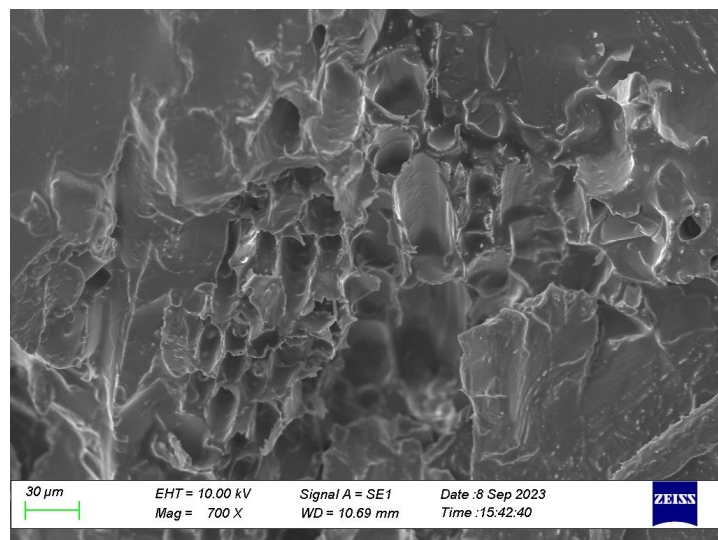


(b)

Fig. 9. SEM images of fractured surface (impact loaded): (a) WPC P-U, (b) WPC T-A.



(a)



(b)

Fig. 10. SEM images of fractured surface: (a) WPC S-U, (b) WPC S-ACA.

3.2.3 Hardness

The hardness of composites is a crucial attribute, mainly when intended for structural applications. The hardness of WPC is influenced by factors such as the type of polymer, the porosity of the wood species, and its density. The hardness value of WPCs generally increased with an increase in filler [39]. In this study, the wood powder content was standardized at 20%, and the polymer matrix type was consistent, which was waste EPS. Therefore, these two factors would not be the determinants of differences in hardness. The porosity of the specimen cross-section and the density of the wood powder become the two key factors determining hardness.

In Fig. 11, it is shown that the surface hardness values of WPC reinforced with pine wood powder and teak wood powder are relatively similar. However, the hardness of WPC with silk tree wood powder reinforcement appears to be lower. The possible cause for this could be the cross-sectional porosity, as observed in the SEM results in Fig. 12. SEM images indicated some microvoids on the surface area of a fractured cross section of WPC reinforced by untreated silk tree wood powder. Therefore, the microvoids will likely explain the decrease in the shore hardness.

4 Conclusions

In conclusion, this research delved into the mechanical properties of Wood Plastic Composites (WPC) reinforced with various wood powders, including pine, teak, and sengon, with different treatment methods. The study revealed that the type of wood powder, treatment processes like alkali treatment, and coupling agent application significantly influence the mechanical performance of WPCs. Notably, WPCs' flexural and impact strengths were found to vary, with pine-based WPCs exhibiting the highest values. Alkali treatment, while enhancing compatibility between wood powder and the polymer matrix, did not consistently improve mechanical properties and, in some cases, even decreased impact strength, attributed to lignin content and poor interfacial bonding.

Furthermore, sequential treatment with alkali and coupling agents showed promise in enhancing WPCs' mechanical properties, particularly in flexural strength. Scanning Electron Microscopy (SEM) analysis provided valuable insights into the fracture surfaces of the specimens, revealing the impact of treatment methods on the interfacial bonding between wood powder and the polymer matrix.

Overall, this research underscores the complex interplay of factors affecting the mechanical performance of WPCs and highlights the need for tailored treatment approaches to optimize their properties for various applications, particularly in the construction industry. Further investigation into the effects of lignin and the development of improved treatment methods could lead to more robust and sustainable wood plastic composites.

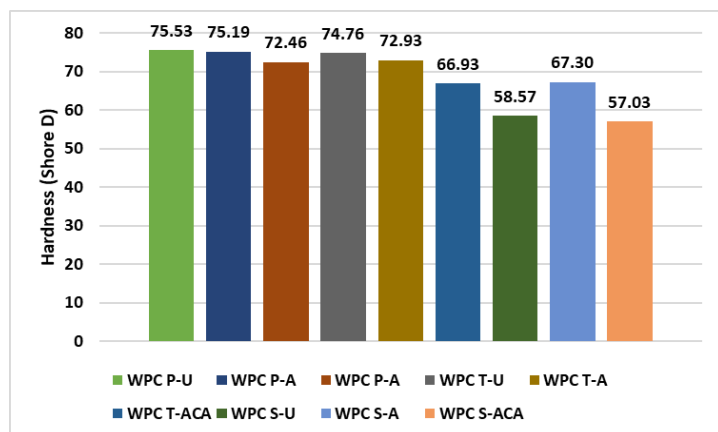


Fig.11. Hardness of WPC.

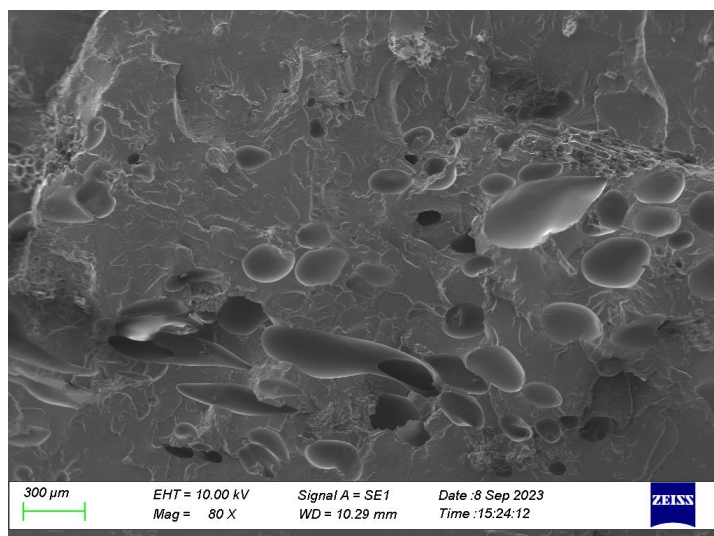


Fig. 12. Cross section of WPC reinforced untreated sengon wood powder.

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