

Characterization of Wood Plastic Composites (WPC) Based on High Density Polyethylene (HDPE), Hardwood Sawdust Residues and Butadiene Rubber (Tyre Dust)

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Abstract: The depletion of natural resources due to the aggressive industrialisation in the last decades has brought considerable attention to research aimed at developing green and sustainable products using eco-friendly materials. Owing to their improved environmental sustainability and the ensuing decrease in carbon footprint, the utilisation of wood fibres to create biocomposites, known as Wood Plastic Composites (WPCs), is a well-researched topic in contemporary scientific research. The objective of the present investigation was to synthesise Wood Plastic Composites using Ground Tyre Rubber (GTR) along with recycled HDPE and sawdust in five different tyre rubber concentrations (1%, 1.5%, 2%, 2.5%, and 3%). The WPCs composite tensile strength and hardness were observed to diminish as the GTR concentration increased, going from 11.9 to 7.63 MPa and 75–70 Shore D, respectively. Additionally, the density was found to increase from 1181 to 1261 kg/m³. For this proportion of GTR, manufacturing conditions have little influence on the composite's final characteristics in terms of strength and applicability. This work provides a reference for further research in related fields.

Key words: Wood plastic composite (WPC), ground tyre rubber (GTR), rubber-wood plastic composite (R-WPC), extrusion, high density polyethylene (HDPE).

Introduction

Industries, driven by concerns for environmental preservation, are increasingly adopting eco-friendly materials and exploring the use of recycled and waste materials. To complement these initiatives, a growing number of research studies have been focusing on utilising less virgin materials and have adopted the use of recycled thermoplastics in the manufacturing of composites made of plastic and wood, which is one of the notable developments in the space (Najafi, 2013).

Plastic wastes, such as high-density polyethylene, are a large component of municipal solid waste, providing recycling challenges for an industry seeking to improve waste utilisation. Large amounts of plastic waste

generated have a significant impact on ecosystems and wildlife, posing a major threat to the habitat and affecting both aquatic animals and human health through fish consumption (Awoyera & Adesina, 2020; Singh & Sharma, 2016). Over the past decade, recycling plastic as a raw material source for WPCs has shown potential due to plastic's vast volume and low cost (Sommerhuber et al., 2015). Moreover, incorporating recycled plastic waste into these composites can replace all solid components, enhancing plastic reuse potential and significantly enhancing environmental sustainability (Awoyera & Adesina, 2020).

Furthermore, the integration of wood fibres for making wood plastic composites (WPC) improves the stiffness and tensile strength of thermoplastics. It

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also utilises considerable amounts of waste generated by the wood industry, which is usually dumped into water bodies and open spaces, or burned outdoors, contributing to environmental contamination and greenhouse gas emissions, affecting both terrestrial and aquatic ecosystems. Additionally, sawdust, a byproduct of wood production, is produced in significant quantities with around 5% created per tree log (Rominiyi et al., 2017). Therefore, reducing the quantity of sawdust generated is crucial for environmental sustainability (Owoyemi et al., 2016).

The increase in rubber waste, particularly in the form of used tyres due to rapid development of the automobile industry, is also a major concern, with significant challenges connected with the recovery and recycling of old tyres due to the very complex structure and composition of tyre components (Luo et al., 2017). Vulcanized rubber further presents a recycling challenge since it cannot be molten or dissolved. Consequently, there is growing interest in finding new ways to recycle or use waste tyre rubber, currently, it is processed into a fine powder-like state for use in areas like outdoor flooring, pavement, sports tracks, and road construction, where there is limited demand and added value (Zhou, 2018).

To create a type of compound that combines materials with both thermoplastic and elastomeric properties, ground tyre rubber (GTR), can be utilised to economically recycle and valorize the discarded tyres. It is widely accepted that the successful incorporation of even a small amount of GTR into thermoplastics would lead to considerable recycling of waste tyres due to the massive market share.

The primary objective of this paper is to explore the characteristics of a composite material derived from recycled High-Density Polyethylene (HDPE), sawdust, and ground tyre rubber. Generally, the integration of tyre rubber into wood plastic composites requires the addition of coupling agents and additives to improve the interfacial bonding. The study aims to seamlessly integrate this composite into the existing manufacturing procedure without making any significant changes to the current process. This would enable manufacturers to incorporate waste rubber readily, consequently enhancing the global recyclability of discarded tyre rubber.

Background

Composite materials are composed of matrix and reinforcement materials. The reinforcing material

supports the stresses inside the composite while a matrix material transfers mechanical stresses. WPC refers to the combination of virgin or recycled plastic with natural fillers like wood flour for reinforcement. The most useful polymers for the manufacture of WPCs include high-density polyethylene (HDPE) and polypropylene (PP). Other recycled polyolefins are also useful, because 140 million tonnes of synthetic polymers (60–65% polyolefins) are produced worldwide every year and residues from these materials are increasing at the rate of 25 tonnes per year (Adhikary et al., 2008a; Fakhrul & Islam, 2013). Using natural fibres like wood dust in reinforced composite technologies has been widely investigated owing to their low cost, high specific properties, renewable nature, and biodegradability (la Pujari, 2013; Engineering Materials, 2021). Reinforcements can be done using different waste materials including timber industry by-products (Kajaks et al., 2014, 2015, 2016, 2017; Kuka et al., 2016). Furthermore, adding wood fibres to plastic products makes good use of waste wood. (Faruk and Bledzki, 2011).

When compared with conventional and mineral filler-reinforced thermoplastics, WPCs exhibit greater strength and modulus, lower density, cost and friction during compounding (Kim and Pal, 2010). In contrast to commonly used wood, WPCs are superior in resisting fungi and termites and provide outstanding dimensional stability when exposed to moisture.

Even though WPCs display superior properties to mineral-reinforced thermoplastics, the moisture present in wood fibres causes undesirable voids in solid WPCs and dimensional variations resulting in poor adhesion due to its sorption to the fibres and consequently lowers the mechanical properties (Schwarzkopf & Burnard, 2016). Using coupling agents during the WPC preparation process is the most practical technique to improve the poor mechanical and physical properties of a composite that are attributed to the weak fibre-matrix interfacial bond (Schwarzkopf & Burnard, 2016; Faruk & Bledzki, 2011). The surface modification of fibres or the use of external processing aids can also facilitate the dispersion and adhesion of these fibres in the polymer matrix resulting in better distribution of the fibres in the polymer matrix while improving all the desired properties (Adhikary et al., 2008b; Faruk & Bledzki, 2012). Moreover, improvement in flame retarding properties of WPCs is obtained using flame retardants, expanding their range of applications.

Based on reactive groups, siloxanes or maleic anhydride derivatives are the most widely used coupling

agents. Some of the most effective interfacial modifiers are maleated polyethylene (MAPE) waxes. The recycling of polymers and wood waste has aroused significant interest in WPCs, leading to quick development over the last three decades (Faruk & Bledzki, 2012).

Literature Review

WPCs started being produced by the plastics industry which had prior expertise in the processing and manufacturing of plastic products (Clemons, 2002). In Europe, WPCs account for nearly 11 % (260,000 tonnes) of composite product production, which includes product categories ranging from construction to consumer goods (Carus et al., 2015). WPCs are considered a growth market, with increases in major markets like North America and Europe estimated to be around 10% while in China growth is estimated to reach 25% in 2015 (Eder and Carus, 2013).

Utilisation of WPCs continuously increases in the world with researchers focusing on studies of different properties of these composites that can be exploited (Dai & Fan 2014, Klyosov, 2007).

Previous studies (Kajaks et al., 2014, 2016, 2017) demonstrated the potential of plywood sanding dust (PSD), a timber industry by-product, in wood-polymer composites (WPCs). Optimal PSD concentrations varied (50 wt-% for HDPE; 40 wt-% for polypropylene), and coupling agents (MAPE and MAPP) enhanced properties, notably impact strength and water resistance, particularly in composites with 40–50 wt-% wood fibre residue (Kajaks et al., 2015). Kazemi Najafi (2013a) delved into the impact of reprocessing cycles on the tensile properties of wood fibre–PP composites, revealing a notable increase in tensile strength by 14% after 2 cycles and decreases in both tensile strength and Young's modulus by 25% & 17% respectively after 8 cycles. D'Almeida et al. (2004) conducted a comparative analysis of flexural properties in WPCs fabricated with recycled and virgin HDPE, underscoring the superior flexural strength and modulus in recycled HDPE-based composites. Bledzki and Sperber (2002) demonstrated the influence of MAH-PP coupling agent on water absorption resulting in improved water resistance. Kamdem et al. (2006) studied pinewood flour-reinforced HDPE composite, achieving a 35% increase in tensile strength and 11% higher impact strength compared to pure polymer. Furthermore, Tisserat et al. (2013) provided insights into the effect of wood flour size on PLA composites using the extrusion method at 170°C, highlighting the impact of an increase

in tensile modulus by ASTM standard with decreasing size of reinforcement.

This study will provide information on the potential use of sawdust residues, which are abundant in India, in the development of WPCs based on recycled HDPE and ground tyre rubber through extrusion. In this composite, plastic will ensure that the final product will be light, water-resistant, tightly bonded and uniform while sawdust will give it the necessary structure and rigidity.

Materials and Methodology

Sourcing of Raw Materials

The WPC materials were produced from multi-component formulations, consisting of sawdust, thermoplastic, ground tyre rubber, inorganics and process additives. Sawdust was sourced from sawmills locally. High-density polyethylene (HDPE) pellets were used for thermoplastic, along with additives and inorganic components such as PE wax, UV stabiliser, antioxidant and calcium carbonate which modified the mechanical and fire retarding properties of the R-WPC. Process additives were also added. The recycled ground tyre rubber was obtained from scrap/used tyres at an automobile repair shop near Okhla which was cut into small pieces using a cutter and then shredded.

Synthesis of WPCs

For manufacturing WPCs into usable products, critical components of the composite, including wood dust, polymer, and additives, are combined in a process called compounding. The process is undertaken in a hopper to impart the desired distribution. It is immensely critical to disperse the wood particles evenly throughout the molten polymer, especially in high-filler WPCs (Schirp & Stender, 2009). A homogeneous mixture of wood dust, additives and molten polymer is prepared to produce pellets. Ensuring a uniform dispersion and encapsulation of all these components within the polymer is essential for imparting optimal mechanical properties to the final composite. In this way, maximum durability and load capacity will be achieved with the composite. The pellets obtained were then passed through a grinder after mixing with recycled tyre rubber to obtain a powdered mixture. Using a counter-rotating conical single-screw extruder, the mixture is processed into the resulting WPC samples in this study. A slit die attached to the extruder, cooled to below room temperature (20°C) using water, gave the final shape to the WPC samples. The process duration for a 10-kg batch of each WPC formulation was 45 minutes.

Six samples were obtained with different tyre rubber and pellet percentages. The pellet composition is given in Table 1 and the formulation of samples is given in Table 2.

Table 1: Composition of wood plastic pellet

<i>Material</i>	<i>Percentage (%)</i>
Recycled HDPE	25
Sawdust	56.82
Calcium Carbonate	15.91
Other Additives*	2.27

*PE Wax, UV Stabiliser & Antioxidant

Table 2: Composition of different rubber, wood and plastic composites

<i>Composite</i>	<i>Pellet (%)</i>	<i>GTR (%)</i>
Baseline WPC	100	0
1% R-WPC	99	1
1.5 % R-WPC	98.5	1.5
2% R-WPC	98	2
2.5% R-WPC	97.5	2.5
3% R-WPC	97	3

Testing Procedure

Tensile Testing

Testing was done according to IS 3400-1:2021 using the Universal Testing Machine (Zwick/Roell; Model: Z010, USA). Six dumbbell-shaped specimens of the WPC with rubber were subjected to testing. Specimen thickness was measured both at the centre and at each end using a vernier caliper. The median of the measurements was employed to determine the cross-sectional area. The resulting curves were plotted.

Density Testing

Density tests were conducted on a Shimadzu Uni Bloc balance (Model: AUX220) according to IS 3400-9:2020. Test sample mass was calculated, followed by its immersion in water, using the water displacement method to calculate the volume of the specimens, which was then used to determine the density values.

Hardness Testing

Hardness measurements were taken following the guidelines outlined in ISO 48-4:2018(E). The measurements were taken by utilising a durometer (Shore D scale; Milhard; Model: DA-2). Three measurements were taken 6 mm apart for each sample, and an average of these measurements has been reported in this study.

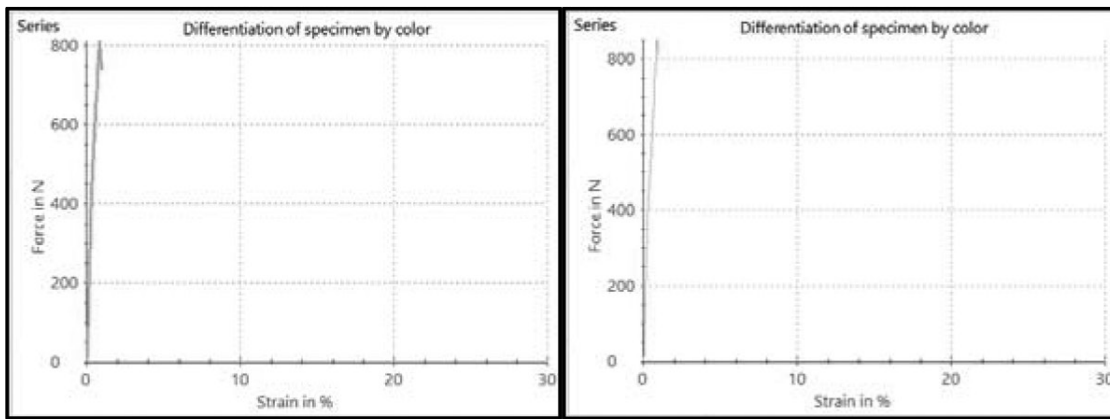
Results and Discussions

Table 3 outlines the key physical and mechanical properties of the WPCs observed during the testing of the six composite samples, including the baseline WPC. The values obtained for tensile strength, density, hardness and elongation at break were determined.

Figure 1 shows the curves obtained during tensile testing of different composites produced. From the tensile strength results, Composite 1 (Baseline WPC) has the highest tensile strength amongst the composites while 3% R-WPC has the lowest tensile strength. The tensile strength for the composite samples was found to vary between 7.63 to 11.9 MPa. Figures 2 and 3 exhibit the density and hardness of WPC composites with varying GTR concentrations along with baseline WPC. The density of composite samples increased with the increase in GTR proportion spanning in the range of 1181 - 1261 kg/m³. Whereas the increase in GTR concentration reduced the hardness of the WPC composites. Furthermore, prior research investigations have corroborated the findings of this study by demonstrating that GTR proportions impair composite

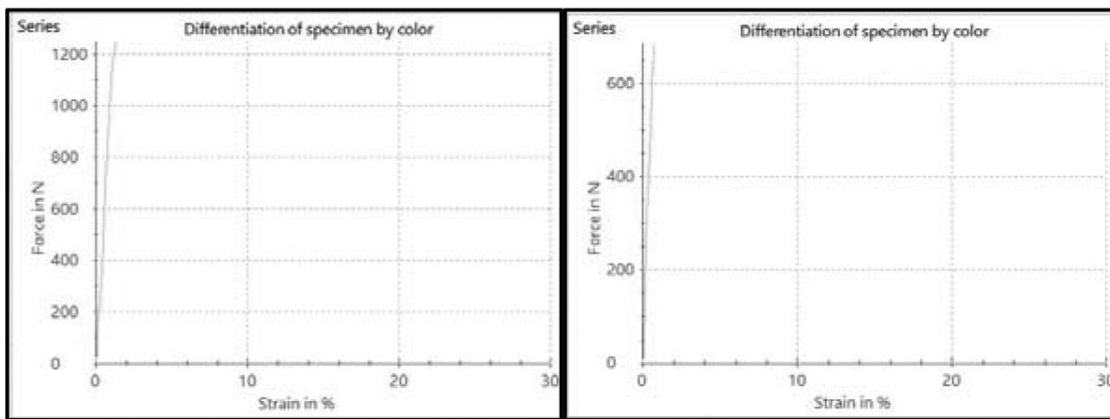
Table 3: Tensile strength, density, hardness and elongation at break for each composite prepared

<i>Composite</i>	<i>Tensile strength (10⁶ N/m²)</i>	<i>Density (kg/m³)</i>	<i>Hardness (Shore D)</i>	<i>Elongation at break (%)</i>
Baseline WPC	11.9	1181	75	1.0
1% R-WPC	11.6	1236	73	1.0
1.5 % R-WPC	10.0	1254	71	0.9
2% R-WPC	9.47	1251	71	0.7
2.5% R-WPC	7.72	1259	70	0.7
3% R-WPC	7.63	1261	70	2.4



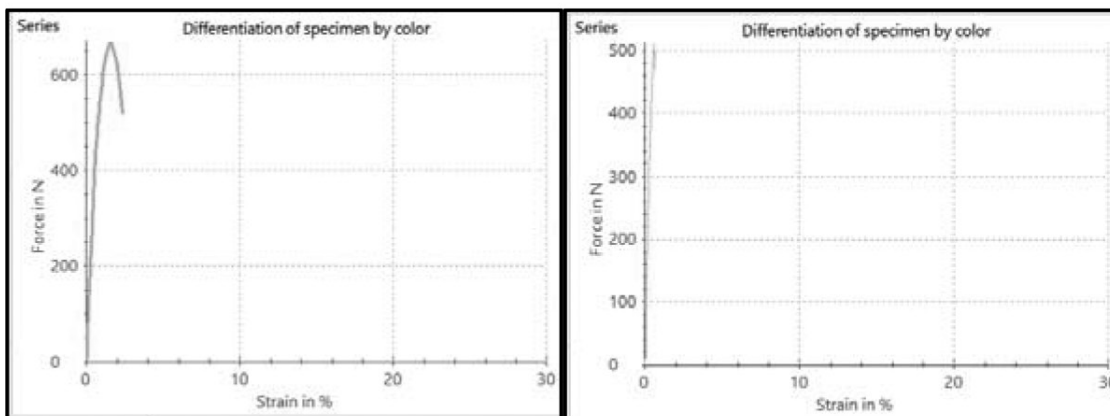
(a)

(b)



(c)

(d)



(e)

(f)

Figure 1: Graphs obtained during tensile testing of (a) Baseline WPC (b) 1% R-WPC (c) 1.5% R-WPC (d) 2% R-WPC (e) 2.5% R-WPC (f) 3% R-WPC.

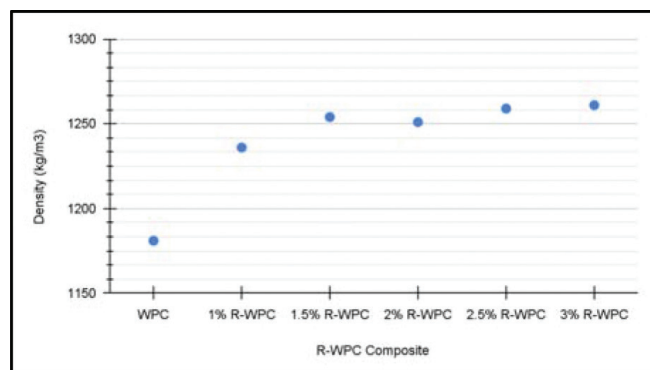


Figure 2: Density variation in WPCs with different rubber composition

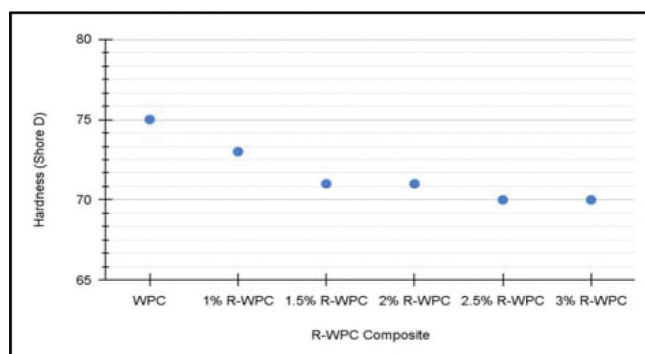


Figure 3: Hardness variation in WPCs with different rubber compositions.

tensile strength and stiffness because rubber has lower mechanical properties than HDPE and does not appreciably toughen the material.

The recycling rate of used tyres is only about 65%, and there are nearly 950,000 tons of non-biodegradable waste rubber, leading to black pollution (Chen et al., 2017). Adding ground tyre rubber to natural fibre-reinforced polymer composites has shown to be a promising avenue for recycling waste tyre rubber. The global volume of tyre rubber is increasing at a massive pace with a projected growth of 6.6 million tons by 2024, indicating a compound annual growth rate of 10.6% during 2019–2024 (Mazzanti et al., 2020).

This research discusses the possibilities for reducing tyre rubber by incorporating it in wood-plastic composites. The study examines the effects of adding up to 3% tyre rubber in the traditional WPC manufacturing process, without the need for additional additives.

Conclusion

In this study, WPC composites with ground tyre (GTR) rubber were investigated. Six different percentages of GTR were added to form the composites under the same processing conditions.

The study revealed that the addition of ground tyre rubber in the WPC composite decreased the tensile strength of the composite. Further, it was observed during testing that Baseline WPC displayed a tensile strength of 11.9 MPa, the highest among all 6 samples, whereas the 3% R-WPC sample recorded the lowest tensile strength at 7.63 MPa. A decreasing trend in hardness was noticed with the increasing addition of GTR in the composite mixture, the highest hardness recorded was 75 shore D for baseline WPC, which subsequently was brought down to 70 shore D as the rubber concentration increased in 3% R-WPC composite. The study

successfully concludes that the utilisation of rubber in the manufacturing of WPC holds a bearing on the composite's mechanical as well as physical properties to a certain extent but is not significant enough to discourage its usage in industry-wide applications.

This study aimed to have an exploratory investigation into the changes brought on by the use of ground tyre rubber as filler material in WPC. This study provided strong evidence in favour of using GTR in WPCs to increase the recyclability of tyre rubber, enabling the transition to a circular economy.

Properties like morphology, water absorption, and flexural strength of the recycled WPCs need to be further investigated in order to assess the composites' suitability for various applications, including decking, railing, and fences.

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