Recycling of Wood – Plastic Composite Prepared from Poly (Ethylene Terephthalate) and Wood Sawdust

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\textbf{A B S T R A C T}

Plastic waste has become one of the humanities and the ecosystem balance serious environmental Challenges. Furthermore, it is the primary source of plastic pollution because it is inexpensive, widely available, and frequently discarded. Using various waste materials and side fractions as part of wood-plastic composites is one way to promote the circular economy (WPC). Several environmental benefits can be realized by using recycled plastic, including extending the usable life of plastic, reducing waste, contributing to the development of trash recycling, and preventing resource depletion. One of the most efficient recycling processes is glycolysis; the (PET) is depolymerized by ethylene glycol in continuous stirring reactors at temperatures between 200 and 220°C using glycol as solvent. This work concentrates on the experimental investigation of composite materials from DE polymerization PET, Unsaturated polyester, and VV/55 as a matrix and wood sawdust as reinforcement. The composite samples were checked by the Hardness test, water test, and density test. According to the experimental results, the optimum value is at (2\%) wood percentage, giving high hardness value, low density, and low water absorption.

\textbf{1. Introduction}

A composite material made of recycled plastic and wood waste is a wood–plastic composite that is frequently abbreviated as WPC. On the market, use other applications use purely virgin raw materials. Its most common application is in the flooring of outdoor decks. However, it can also be used for railings, fences, landscaping timbers, cladding and siding, park benches, molding and trim, window and door frames, and indoor furniture. According to the manufacturers, WPC is more environmentally friendly and does not require much maintenance compared to hardwood treated with preservatives or solid wood of the rot-resistant species. These materials are resistant to breaking and splitting and can be molded with or without simulated wood grain characteristics [1, 2]. WPC are appealing die to their manufacturing process is highly automated and adaptable to a wide range of species and raw ingredients. Improvements in processing technology, the development of acceptable chemical coupling agents, and economic factors have all aided the usage of wood in thermoplastics. The rising operating temperatures, cycle times, and mold shrinkage have a major contribution to the rise of the fiber/plastic composite industry [1]. Plastic materials are widely employed due to their numerous benefits, including low density, ease of processing, corrosion resistance, durability, low cost, and high strength [3]. Plastic including nylon (PA), polyethylene (PE), polystyrene (PS), and polyethylene terephthalate (PET) are widely used in WPC manufacturing [3, 4]. Using various waste materials and side fractions as part of wood-plastic composites is one way to improve the circular economy. Due to extensive breakdown durations, the accumulation of plastic trash generates a variety of environmental problems, various production processes generate a considerable amount of waste materials, and solid waste management is a major environmental issue around the world. As a result, converting plastic trash into value-added products has emerged as a viable option for boosting the world economy from plastic waste [3, 5]. Wood, as a natural renewable material of biological origin, has numerous advantages in a variety of applications, ranging from structural and construction materials to furniture and transportation to raw materials for...
energy production, due to its unique microscopic structure, abundance, relative ease of work, and good visual effect [6]. Only a few of the advantages of wood-sawdust over other typical wood composite materials include better insulating qualities, resistance to water absorption, fire performance, and strength properties. Wood-sawdust can save money on materials, repurpose wood waste, and provide equal thermal performance and strength. Building using wood-sawdust with structural support is possible, and it is especially advantageous in poorer nations due to its inexpensive cost [4]. Hemicelluloses, starches, sugars, phenols, and hydroxylase carboxylic acids are among the compounds in wood that might dissolve and alter lime crystallization. The hydrophilic property of the cellulose structure of the fiber is linked to the major difficulties of wood composites. Using an alkali solution, partially remove sawdust components such as cellulose, hemicellulose, lignin (the major components that contribute to the strength, flexural, and impact qualities of composites), pectin, waxes, and water-soluble compounds. The explanation for matrix cohesiveness with treated sawdust is that as lignin is removed, the middle lamella connecting the final cells becomes more flexible and uniform as micro-voids are gradually eliminated[4,7]. According to the researchers, the hydrogen bonding in the network structure is disturbed after alkaline treatment, enhancing surface roughness. The procedure depolymerizes cellulose and exposes the short-length crystallites by removing a layer of lignin wax and oils from the fiber cell wall’s exterior surface. The bonding between sawdust and hydrophobic matrix has a direct impact on the mechanical properties of composite materials, while studies have demonstrated that removing hemicelluloses results in a less dense and rigid inter-fibrillary area [8, 9]. This project aims to create a wood-plastic composite (WPC) material from depolymerized polyethylene terephthalate (DPET), unsaturated polyester (UPS) as the matrix, as well as wood sawdust as reinforcement for use in construction application, partition board.

2. Experimental work

The experimental work consists of three sections:

1) The first section is the prepared PET powder from bottled water waste
2) The second section is prepared Wood-Sawdust (grinding, sieved to a size of (54 µm-36 µm), alkali treatment).
3) The third section is the prepared composite material by incorporating DPET powder with VV/55, UPS as a matrix, and wood-sawdust as reinforcement. Four different percentages of (1, 2, 3, 5 %) per weight of wood are used.

2.1 PET powder Preparation

Polyethylene terephthalate (PET) Waste from water bottles was gathered from trash bins in Baghdad, Iraq. A lab grinder and an industrial mill were used to granulate the bottles gathered. DPET is produced by the DE polymerization reaction, which can be broken down the pet into monomers, oligomers, and other chemical components by chemical recycling process; Glycolysis reaction with glycol in the presence of Tran’s esterification catalysts (Nano MgO) is used to accomplish this. After that, the DPET polymer powder is washed with distilled water for an hour and then nominated using nomination paper and left to dry [10-13]. Table1 shows the properties of DPET [14] and Table 2 shows the properties of Nano MgO and ethylene.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>1.38 g/cm³ (20 °C)</td>
</tr>
<tr>
<td>Melting point</td>
<td>260 °C</td>
</tr>
<tr>
<td>Molar weight</td>
<td>192.2 g/mol</td>
</tr>
</tbody>
</table>

Table 2: Main properties of ethylene and magnesium oxide

<table>
<thead>
<tr>
<th>Materials</th>
<th>Origin</th>
<th>Purit%</th>
<th>Grain size</th>
<th>Total projected area (rel.)</th>
<th>Specific surface area(SA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethylene Glycol (EG)</td>
<td>Ethylene Glycol (EG)</td>
<td>99.99%</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Nano MgO</td>
<td>Sky Spring, Nanomaterial's, (USA)</td>
<td>99%</td>
<td>60 nm</td>
<td>14.65%</td>
<td>50</td>
</tr>
</tbody>
</table>

The unsaturated polyester resin which is a thermoset polymer (produced by Saudi industries resin limited, Jeddah, Saudi Arabia) used with DPET as a matrix. The hardener A Methyl Ethyl Ketone Peroxide hardener is used to transform it from a liquid to a solid-state. Ravels VV/55 As a binder, it’s an anionic aqueous dispersion of vinyl acetate copolymer with a versa tic ester with shallow residual content. VISICRYL828 is an aqueous APEO& FORMALDEHYDE FREE Acidic cross-linked acrylic thickener for water-based paint, textile flocking, and printing and printing ink production applications. Until neutralized with alkali to pH 8, it will convert into a very viscosity gel-like material. Application of VV/55 used Indoor and outdoor water-based stain and glossy paints, textured coatings, primers. Ravels VV/55 is distinguished by an optimal blend of several aspects affecting product performance in the construction sector: the power of binding, rheological behavior, film sensitivity of the water, compatibility with a variety of fillers, pigments, and additives, resistance to atmospheric agents. Ravels 55/VV-based
exterior water-based paints have low water absorption and good vapor permeability. Table 3 shows the main properties of Ravels 55/VV.

Table 3: The main properties of Ravels 55/VV

<table>
<thead>
<tr>
<th>Supply specifications.</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solid content</td>
<td>%</td>
<td>55±1</td>
</tr>
<tr>
<td>Brookfield viscosity</td>
<td>Mpa.s</td>
<td>9000±2000</td>
</tr>
<tr>
<td>PH</td>
<td>--</td>
<td>4.5±0.5</td>
</tr>
<tr>
<td>Prevailing particle size range</td>
<td>µm</td>
<td>0.2-2.0</td>
</tr>
<tr>
<td>Dispersing system</td>
<td>--</td>
<td>Anionic – non-ionic</td>
</tr>
<tr>
<td>Density at 23°C</td>
<td>Kg/dm³</td>
<td>1.1</td>
</tr>
</tbody>
</table>

2.2 Wood –Sawdust preparation

Wood sawdust White Cham with density (0.21 gm/ cm³), type (LE), (originally from Iraq) obtained from local wood carpentry workshop was used in this work. A batch of sawdust was sieved using a sieves shaker (FRITSCH –Germany to a size of (54 µm-36 µm), located in the laboratories of the Materials Engineering Department in University of Technology. Distilled water was used to wash the sawdust with a ratio (5 gm/ 5 l) for half an hour at the heat of the cooker. After washing completion, the solution was filtered, and the fibers were dried under the effect of sun rays for 72 hours, and then dried in an oven at 80 °C for 3 hours. Figure 1 shows sawdust used in this research.

![Figure 1: (a): wood sawdust from the workshop (b): wood sawdust after grinding and washing and (c): wood sawdust after alkali treatment at room temperature.](image)

The natural powder (wood sawdust) was soaked in an alkali solution consisting of 6% (w/v) of (NaOH) in distilled water, at room temperature (25 °C). Sawdust powder was soaked in sodium hydroxide solution at different concentrations room temperature for 3 hours [15]. For instance, to make a 6% NaOH solution, a 60 g NaOH pellet was dissolved in 1 liter of distilled water. After that, the treated powder was rinsed multiple times with distilled water to eliminate any leftover (NaOH) on the surface until it reached a neutral pH of 7. Subsequently, the powder was allowed to dry at room temperature for three days before being placed in a furnace at 30 °C to ensure thorough drying.

2.3 Preparation of composites specimens

The amount of reinforced material (sawdust) is weighed using an electronic balance with an accuracy of (0.0001) digits according to the required selection ratio of the weight parts of the reinforcing material, based on the total weight of the matrix material required to fill the mold cavities. The matrix in this research consists of three materials (DPET, VV/55, and UPS), at room temperature, homogenous and continuous mixing of liquid UPS resins with DPET is required. After that, added VV/55. As a result, it must ensure that the mixture is homogeneous. Depending on the compatibility between matrix materials and several mixing ratios, the perfect matrix ratio is [50% DPET+20% VV/55, UPS is Balance]. The wood sawdust powder was then added to the mixture, and the Sawdust was progressively mixed in to create composite materials, the composite was poured into the mold and opened after two weeks at room temperature to solidify it, this process is important to reveal the complete chemical reaction between materials. The Composite samples were ripped off from the silicone Mold cavities with the very smooth upper and lower surface. Table 4 shows the concentration of the reinforcement materials for all specimens of these composite.

Table 4: DPET composite specimens and the reinforcement composite specimens that preparations in this research

<table>
<thead>
<tr>
<th>Composite number</th>
<th>UPS %</th>
<th>Sawdust % (treated/untreated)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>29</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>28</td>
<td>2</td>
</tr>
<tr>
<td>3</td>
<td>27</td>
<td>3</td>
</tr>
</tbody>
</table>
3. Characterization

3.1 Mechanical and Physical Test

The effects of the WPC industrial scrap mix ratios and the number of recycling passes on the DPET/wood flour WPC were evaluated using the mechanical tests below:

3.2 Tensile test

Tensile samples and tests are carried out according to the standard specification of ASTM D-638 by using the global tensile device type (LARYEE) [16]. Figure 2 illustrates the sample for the tensile test which is standard and a sample of the composite.

![Figure 2: (a) the standard specimens (b) Experimental Tensile Test Specimen before Testing and after testing men for the tensile test](image)

3.3 Hardness test

Hardness is a property of a solid substance that describes its resistance to penetration, wear, and scratching on its surface. The hardness value can be used to estimate the WPC composite materials’ strength, structure coherence. Temperature, intermolecular connections, a structure such as crosslinks in chains, volume fraction of particles reinforcing, and particle size all influence the hardness test [17]. The hardness test was carried out at room temperature using the Documenter 3120 type is (Shore D), manufactured in the USA. Hardness test apparatus in accordance with (ASTM D2240), the applied load was nearly equal to the 50 N. The depressing time of measuring was nearly equal to the (5sec). All the specimens which had dimensions 10 mm thickness and 10 mm diameter were tested 5 times at different positions at the same time and the average values were taken [18]. Figure 3 illustrates the standard specimen for hardness, experimental test specimen during testing hardness device type (Shore-D).

![Figure 3: (a) Schematic for Standard Hardness Test Specimen and (b) Experimental Test Specimen inside Hardness Test Device](image)

3.4 Water absorption test

The material is subjected to deterioration, internal tensions, and dimension instability as a result of water absorption. This results in the production of cracks and fractures in the partition board application [19]. The water absorption test was carried out in accordance with the manufacturer’s instructions (ASTM D1037-99). The dimensions of specimens are 5mm width, 10mm length, and 5mm thickness. These specimens were immersed in distilled water at room temperature for a specified
duration in this test. The specimens were taken out of the water at each testing period, wiped using tissue paper to remove surface water, and weighed using a (Precision balance) produced by (Redwing), type (PS 360/C/1). After entire immersion in demineralized water at a temperature of 20 °C (±5), the water absorption of the composite was determined by differential weighing of the specimens for each time interval (at Days 1, 24 h) to the Eq.1 [19]:

\[
\text{Water absorption} \% = \frac{(W_t-W_o)}{(W_o)} \times 100
\]  

(1)

Where \(W_t\) is the specimen weight after a given immersion in water, and \(W_o\) is an oven-dry mass of the specimen weight after constant mass was reached (weight change <0.1% after 24 h). Figure 4 illustrates the standard and experimental specimens for this test.

\[\text{Figure 4: (a) Schematic for Standard Water Absorption Test Specimen and (b) Experimental Water Absorption Test during Testing}\]

3.5 Density test

The density of the composite specimen gives an indication of the lighter WPC composite in this study. In accordance with ASTM D 792, the composite sample was prepared in this test and the weights were measured according to the Archimedes method by using three digits balances (PS360/C/1) device as shown in Figure 5. To calculate the density of composite material in this study the test specimens were weighted in air and weighted during immersed in distilled water at room temperature. The specimen volume should not be less than (1cm³) and the edges, as well as surfaces, must be free from any (oil, grease, and other foreign matter) and smooth [20].

\[\text{Figure 5: Density device}\]

The density of water was assumed to be 1 g/cm³ in order to calculate the density of the specimens according to the following formulas Eq. 2:

\[D = PL \frac{w_1}{w_1 - w_2}\]  

(2)

Where PL: Density of the liquid used.
W1: Specimen weight before immersion.
W2: Soaked immersed specimen weight.
4. Results and discussion

4.1 Tensile test result

The tensile strength of composite specimens reinforced with alkali-treated sawdust with 6% (W/V) is presented in Figure 6 has the highest failure strength than untreated ones and samples without sawdust (matrix). This due to the alkali treatment with 6% NaOH improved the mechanical properties of composites, due to better interfacial adhesion between the sawdust and the matrix occurs, imparting improvement in the mechanical properties of the composites. Cellulose is responsible principally for the strength of sawdust because of its specific properties such as high degree of polymerization, as well as interfacial bonding between the filler and the matrix has significantly improved upon alkali treatment, leading to increase the stress transfer efficiency from the matrix to filler. From Figure 6 the tensile strength decreased gradually with the increase in sawdust percentage, where the highest value was obtained when (2%). On the other hand, the increase of filler content also produced more filler ends. This means that there is considerable stress concentration points taking place by agglomeration of the filler particles and de-wetting of the polymer at interphase aggravates the situation by creating stress concentration points, the poor interfacial bonding causes partially separated micro spaces between sawdust and polymer matrix.

![Figure 6: Tensile strength of composite samples with untreated/treated sawdust](image)

4.2 The hardness results

The results have been shown in Figure 7, the hardness of the composite with (treated/untreated sawdust) increase with sawdust increase, at the optimum percentage of sawdust (treated/untreated) is (2%). The increase due to alkali treatment of wood sawdust, the presence of (OH) from (NaOH) moved to sawdust and work as a functional group cause a good bond between matrix and sawdust, good adhesion between them reflected the hardness result. The explanation for matrix cohesiveness with treated sawdust is that as lignin is removed, the middle lamella connecting the final cells becomes more flexible and uniform as micro-voids are gradually eliminated.

![Figure 7: Hardness of composite samples with untreated/treated sawdust](image)
4.3 The water absorption result

The water absorption values of composite materials with (treated/untreated sawdust) for all samples that were fabricated in the current work are illustrated in Figure 8. It can be observed from the figure that the water absorption increases with an increase in the weight percentage of sawdust (treated& untreated). This is due to the affinity of the sawdust towards the moisture, and also may be due to the high moisture absorption level of natural fibers in the polymer matrix that result from polar hydroxide groups in the sawdust. In general, specimens with untreated sawdust have a higher rate of water absorption than specimens with treated sawdust. This could be attributed to the presence of increasing amount of (-OH) group as a result of higher sawdust content [21].

4.4 The density results

The measurement of density for composite is an essential indicator to know the lighter composite materials [22]. The relationship between the weight percentage of sawdust powder and the density of the specimens is illustrated in Figure 9. It can be seen that the density values for the composite specimens are decreased with increasing the weight percentage of sawdust powder (treated /untreated). The density of the specimens with treated sawdust gives lower values than untreated, decreased with raising the weight fraction of sawdust powder, because of the alkali treatment of sawdust. This is mainly due to the inclusion of long sawdust powder into the composites decreasing the packing, which leads to the disruption of powder distribution and resulting in high void spaces. Apparently, greater void contents yield low density composite. Therefore, the composite become less dense with increased sawdust percentage [23].
5. Conclusions

1) The lowest water absorption and lowest density of composite was obtained when the sawdust was treated; making it suitable in waterproofing materials construction application.

2) Using the DPET in fabricated blends and composite materials considering reuse and recycling of plastic waste, contributes to protecting the environment.

3) The fabricated composite used in construction applications such as partition boards because of high tensile properties.

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Author contribution

All authors contributed equally to this work.

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Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

References


