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THE UTILISABILITY OF HORNBEAM WOOD SAWDUST WITH CALCITE FILLER IN POLYESTER-BASED COMPOSITE MATERIALS

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Abstract: *Fillers are used extensively in all types of polymers such as thermoplastics, elastomers and thermosets. It is considered impossible to develop many polymers without fillers. The primary purpose of using fillers in polymer materials is to reduce cost, but as polymers have become cheaper and more commoditized today, the main purpose of using fillers is no longer to reduce cost. A wide variety of fillers are available, and with their careful selection, composite materials with improved properties for functional use can be produced. The main reasons for using particulate fillers today vary by polymer type but include improved processing, increased hardness, thermal distortion temperature and creep resistance, better wear and tear resistance, and flame retardancy.*

In this study, it was aimed to use organic-based hornbeam wood sawdust together with inorganic-based calcite filler. Thus, it is aimed to obtain an environmentally friendly green material. General purpose polyester resin was chosen as the matrix material for the composite due to its extensive use in industry. Hornbeam wood sawdust and calcite were used as fillers, mixed with polyester resin in certain proportions. Composite samples were obtained by open mould casting method. The filler rate was determined as 20% limit value, taking into account literature studies. Izod impact tests, tensile tests, and hardness tests of the composite samples were performed and SEM images of the broken surfaces were examined. Since the composite samples contained organic fillers, water absorption tests were also performed. As a result of the study, it was evaluated that the use of hornbeam wood sawdust and calcite together increased the impact and tensile strength, and therefore hornbeam wood sawdust could be used as a filler together with calcite in sectors such as paint and plastic.

Key words: Calcite, green composite, hornbeam wood sawdust, thermosetting polyester

1. INTRODUCTION

Composite materials consist of at least two materials: matrix and reinforcement/filler. The use of reinforcement is mostly used to increase strength. Fillers are powdered substances with low aspect ratio particles, usually micron in size, that are generally added to polymers to reduce cost, improve processing, and/or modify one or more properties.

Particle-based fillers are used in many different sectors and fields such as vehicle tires, printed circuit boards, construction, transportation and cable. It is estimated that 25-30 million tons of particle fillers are used in the polymer industry worldwide. The main purpose of use of fillers is to reduce cost. The main fillers used for polymer-based composites are natural carbonates (calcium carbonates), carbon blacks, kaolin, talc, aluminium hydroxide, precipitated silica and wood sawdust.

In polymeric materials, there is no ideal polymer or filler. Therefore, the choice of fillers is made to maximize certain properties and reduce the loss of other properties and blend the materials. The main purpose when choosing particle-based fillers is to expect high performance and low cost from the material. It has been reported that there are at least 30 properties that can be changed in the structure of the polymer by adding fillers. So you might be adding a filler to increase hardness, but it's clear that many of the other properties would likely be changed in a negative way, even though that wasn't the intent.

The cost of fillers varies greatly, from raw clays and carbonates to special synthetic types. This difference can be up to 100 times. There are large price differences even for a type of filler such as calcium carbonate, depending on factors such as colour, purity, particle size and shape, and surface treatment. For very low-cost fillers, transportation costs have now become important. With the increase in globalization, uniformity of filling qualities around the world has now become an important issue. Additionally, factors other than raw material costs should also be taken into account when evaluating whether cost savings can be achieved. One of the most important of these is the additional transaction costs incurred.

When considering potential cost savings from the use of fillers, it should always be borne in mind that they are often significantly denser than the parent polymer. While the densities of commonly used mineral fillers are in the range of 1.5–3.0 g/cm^3, the density of commercial plastics is in the range of 0.9–1.4 g/cm^3, which means that fillers generally have a density-increasing effect.

It is thought that green composites will make a significant contribution to the environmental problem in the 21st century, as they are a potential solution to the waste problems caused by traditional petroleum-derived polymers. Green composites, which are called environmentally friendly polymeric composite materials, offer techno-economic advantages in engineering applications and also have a very high pressure from the global market. For this reason, intensive studies have been carried out in the literature on green composites recently. In this study, hornbeam sawdust was used as a green composite component.

This tree species, which grows in many parts of the world, was chosen because it is common in our country, especially in the Black Sea region, and it is thought that the wastes from these tree species can be used other than incineration and will benefit the country's economy and energy recovery. The density of the hornbeam tree is 0.83 g/cm^3 and it is quite dense and hard compared to other trees. Due to this feature, it is used in shoe making, tools, firewood, flooring, coating, musical instruments, wheels, sports equipment, handles, spindles, cars, windmills, airplane bottoms, etc. It is used for.

In this study, hornbeam tree was chosen as the biomass source and waste hornbeam sawdust generated both from tree felling and furniture making was used as filler. Calcite, another mineral-based filler, is the most commonly used filler type in the industry. The amount of filler by weight was kept as the upper limit at 20%, the mechanical properties of the composite samples with different contents were examined and SEM analysis was performed on the fractured surfaces of the samples. Thus, it is thought that the wastes originating from this type of wood will be evaluated other than incineration and will benefit the country's economy and energy recovery, and will be used together with traditional mineral-based fillers in polymeric composites.

2. METHODS

2.1 Materials

In this study, the unsaturated polyester resin used as the matrix, general purpose unsaturated polyester resin. Methyl ethyl ketone peroxide (MEK-p) was used as a hardener. Hornbeam sawdust was obtained as coarse sawdust from local furniture manufacturing companies. Sawdust size is in the range of 1-200 microns, and larger and coarser particles are not used. Calcite (calcium carbonate) was obtained from Nidaş Mining and is 3 microns in size. In order to prepare test samples, open casting was made in a Teflon mould. The reason for using Teflon mould is that after the resin hardens, it is easily separated from the mould due to its hydrophobic surface feature.

2.2 Sample preparation

The curing process was carried out at room temperature and 1.5% by weight of 6% Cobalt naphthalate was added to the resin as an accelerator. The mixture was thoroughly mixed until it became homogeneous. Then, 1.5% MEK-p was added as hardener. Hardener was added to the filler added samples after the filler was added. The whole mixture was thoroughly mixed for 2-3 minutes until it became homogeneous. Addition ratios of fillers by weight are given in Table 1.

Table 1: Contents of composite samples (by weight, %)

Samples	Calcium Carbonate	Hornbeam sawdust	Polyester
Neat Polyester	0	0	100
5CC15HS	5	15	80
10CC10HS	10	10	80
15CC5HS	15	5	80
20CC	20	0	80
20HS	0	20	80

Abbreviations: **HS: Hornbeam sawdust; **CC:** calcium carbonate.*

2.3 Mechanical tests

The tensile test of the samples prepared according to the ISO 527 standard was carried out in the Zwick Z010 universal tensile device with a tensile speed of 5 mm/min. The impact strength of the unnotched samples prepared according to the ISO 180 standard was tested using a 5.4 J Izod hammer in the Zwick B5113.30 impact device. Hardness measurements were made in Zwick Shore D device, 15 sec. made with a waiting period. For scanning electron microscope (SEM) analysis, the samples were coated with a 10 Å thick gold/palladium alloy. The SEM test was performed with the Polaron SC branded device located in the Marmara University Faculty of Technology Laboratory.

3. RESULTS AND DISCUSSIONS

In this study, polyester resin, which is the most used in composites, was used as the matrix material, calcite filler, which is also the most preferred inorganic filler, and sawdust of the hornbeam tree, which is abundant in Turkey, was used as the organic filler. Green composite was made by using inorganic and organic filler together and their usability was evaluated. The total filler rate in the matrix was determined as 20% by weight.

In general, inorganic fillers increase the mechanical properties of polymeric composites with the filler content until the optimum value and then decrease. The mechanical properties of filler-based polymers are affected by the interfacial adhesion between the matrix and the filler. Factors such as the filler ratio and size used in the composite and the bond strength of the matrix layer between the fillers also affect the strength. The tensile strength values of the samples are given in Figure 1.

Among the samples, the highest tensile strength was found in the 20 CC coded sample (24,2 MPa), and the lowest tensile strength was determined in the 20 HS coded sample (16,8 MPa). The tensile strength decreased as the HS content increased in the samples. These results are similar to the literature. In addition, it has been stated in many studies that as the amount of organic filler in the matrix increases, the tendency to agglomerate causes the tensile strength to decrease. The tensile strength of neat polyester was determined as 23,1 MPa.

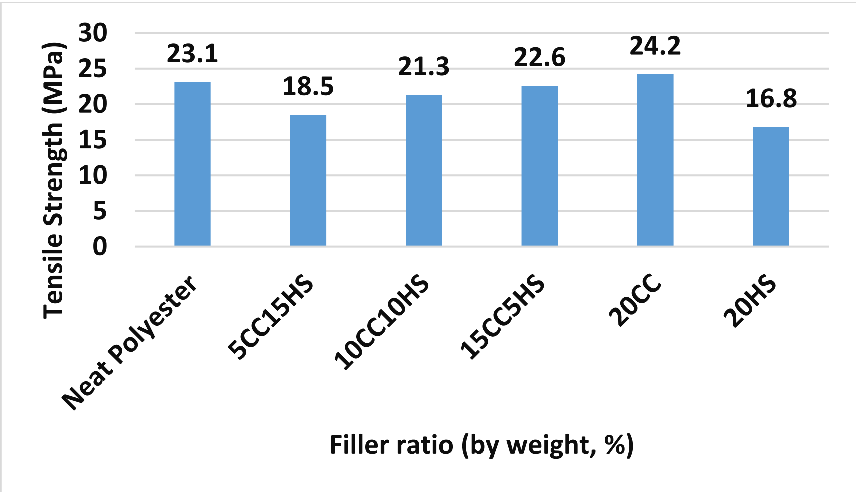

Figure 1: The tensile strength values of the samples depending on the filler ratio

Figure 2 shows the Izod impact strength properties of the samples depending on the filler ratio. The impact strength of composite samples may vary depending on many situations such as the properties of the filler material, the distribution and density of the filler, their interaction with the matrix material, composite production technique, and environmental factors affecting the filling material. Organic fillers can generally be in fibre or particulate form. Long fibres or larger particles generally provide better impact strength because they provide more opportunity to dissipate and absorb energy. However, this relationship is not always true and in some cases large filler sizes can cause undesirable results. Again, the homogeneous distribution of the filler material in the structure and its appropriate density also positively affects the impact resistance.

As expected in this study, a decrease in the impact resistance of the composite was observed with the increase in organic filler ratio, and a partial increase was observed with the increase in inorganic filler. The impact strength of the sample coded 15CC5HS increased compared to pure polyester. The highest impact strength was obtained in the 20% calcite filled sample. It can be said that the filler size in the samples, which was in the 1-200 micron size range, did not have a negative effect on the impact strength, because there was no obvious decrease with the addition of organic filler. Even though sawdust filler was used together with calcite, it showed lower impact strength than polyester after 5% filler rate. It is thought that the formation of weak interfacial bonding in the following ratios and the tendency to clump as the amount of filler increases reduce the impact strength.

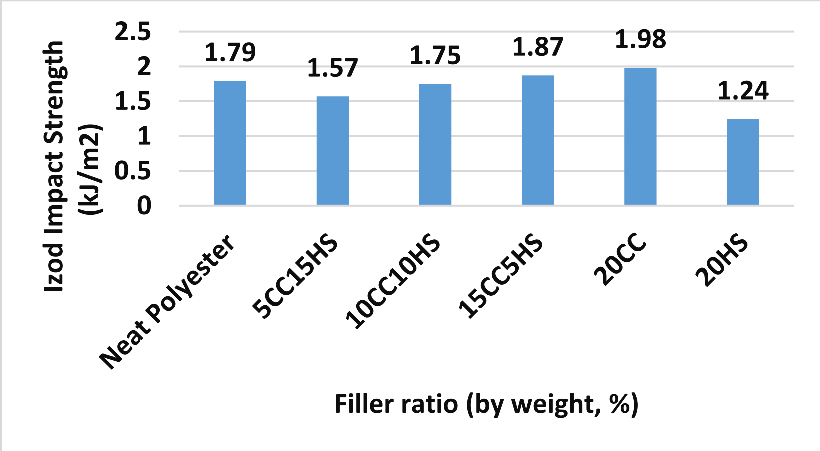

Figure 2: The Izod impact strength properties of the samples depending on the filler ratio

Figure 3 shows the hardness properties of the samples depending on the filler ratio. Inorganic fillers have a positive effect on hardness in composite materials. However, a complex relationship emerges between the size of the organic-based filler and the hardness properties it will provide to the structure. In general, fillers with large dimensions and high-density fillers can contribute to a higher level of hardness, but may vary depending on the filler type, distribution, and other factors. The process of combining organic fillers with the composite material can affect the hardness properties. The pressing processes used in the composite sample preparation stage can affect the homogeneity and therefore the hardness of the composite material. Or, in the open cast production method, the fillings may sink to the bottom due to the density difference. Again, with organic filler materials, moisture, temperature or chemical interactions can affect the properties of the filler material and therefore its hardness properties.

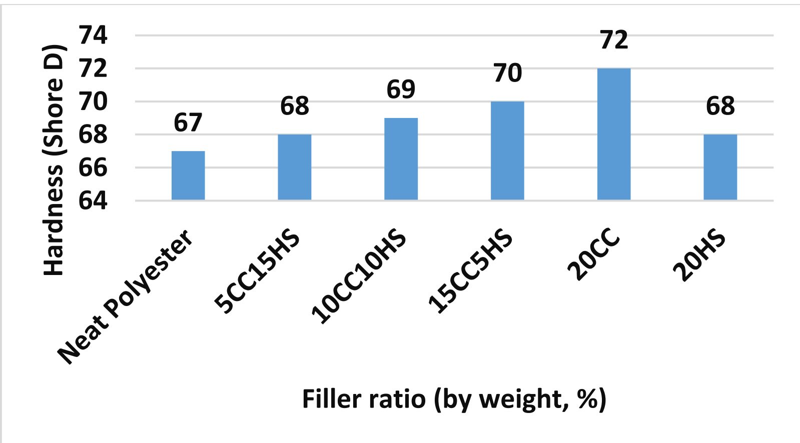

Figure 3: Hardness properties of the samples depending on the filler ratio

Figure 4 shows the density properties of the samples depending on the filler ratio. The density of polymeric composites varies depending on the density and amount of fillers. However, if the bonds between the filler and matrix are strong, the density of the composite can often increase. Smaller particles are generally better dispersed and can form a tighter matrix, which can increase the density of the composite. If the filler particles are homogeneously distributed and have a uniform morphology, the density of the composite generally increases. As expected in this study, the use of inorganic filler amount increased the density, while increasing the amount of organic filler decreased the density.

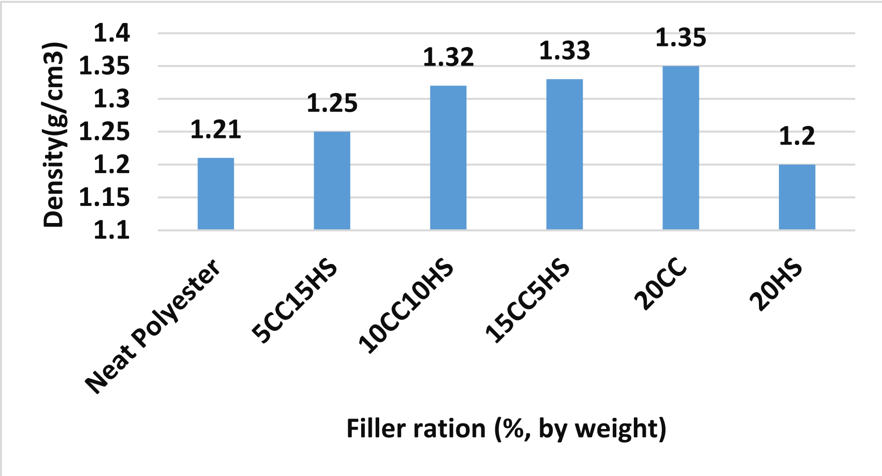

Figure 4: Density properties of the samples depending on the filler ratio

Figure 5 shows the water absorption properties of the samples depending on the filler ratio. Studies in the literature have reported that natural fibres, which are hydrophilic in nature, absorb high moisture and have a rapid water absorption rate. The hydroxyl groups and water molecules of cellulose form hydrogen bonds, increasing the water absorption capacity of the composite. It has been observed that sawdust-filled composites have a similar behaviour. The sample that absorbs the least water is neat polyester. Among the composite samples, the calcite-containing sample is the one that absorbs the least water.

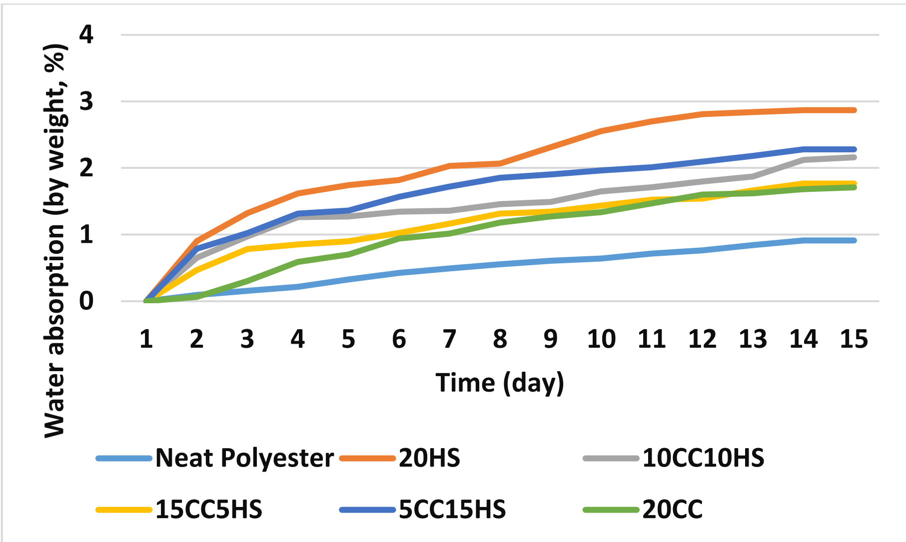

Figure 5: Water absorption properties of the samples depending on the filler ratio

SEM images of the samples are given in Figure 6. In Figure 6a, it is understood that pure polyester is broken by brittle fracture. Figure 6b image contains hornbeam sawdust and calcite filler. It can be said that hornbeam sawdust and calcite filler show a good combination in polyester resin. As can be seen from the images, it is understood that the ends of the sawdust are in the range of approximately 50-100 microns, and the remaining structure is embedded in polyester. It can be said that as the calcite ratio increases in the sample (6c) containing 20% calcite filler, agglomeration occurs. This can cause the formation of bubbles and pores that can interfere with the mechanical properties of the composites. In the 6d image, hornbeam sawdust particles of certain sizes that have protruded from the matrix can be seen. As can be seen from the image, the sawdust particles, which have a cellulosic structure, adhered very well.

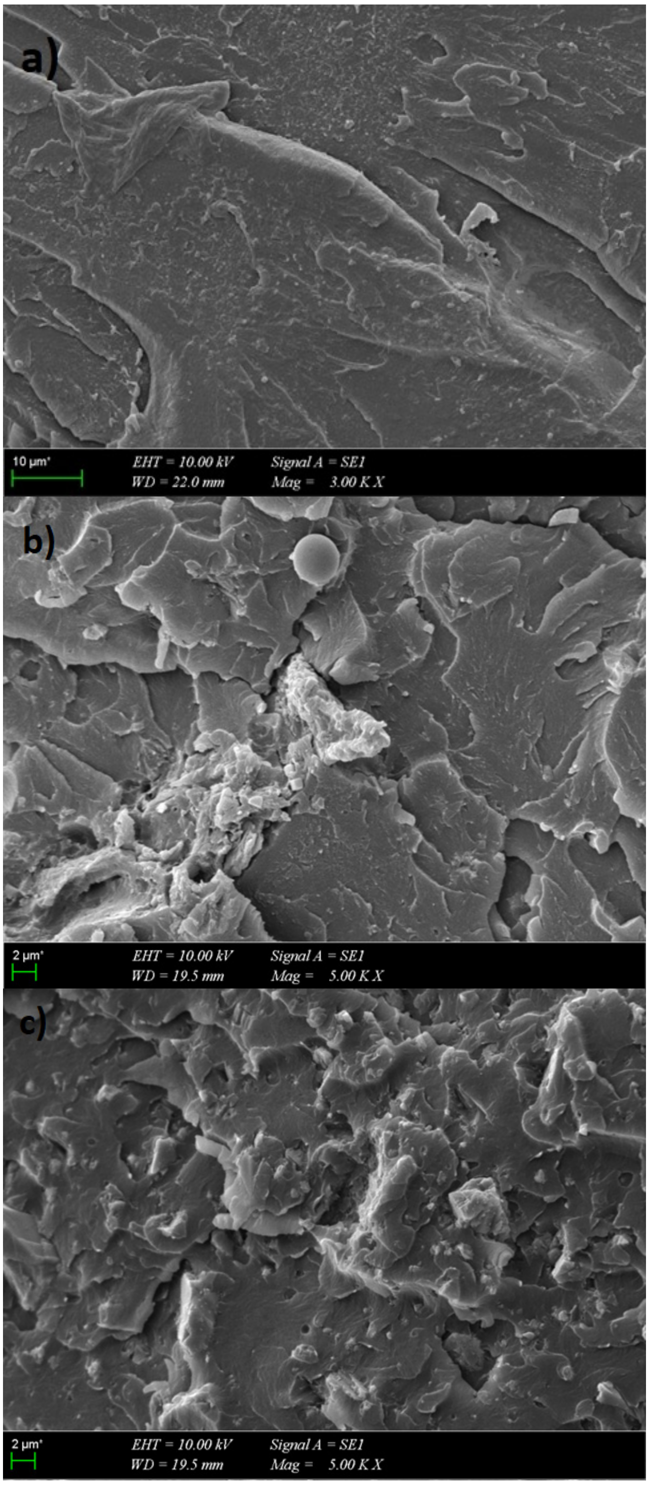

Figure 6 (part 1): SEM images of samples a) Neat polyester b) 10HS10CC c) 20CC d) 20HS

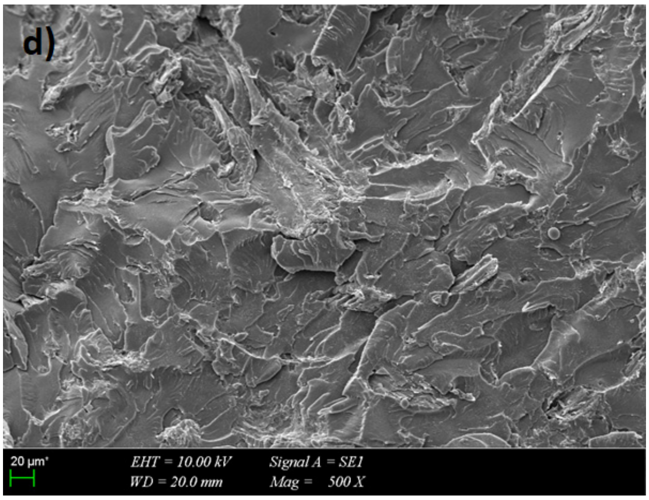

Figure 6 (part 2): SEM images of samples a) Neat polyester b) 10HS10CC c) 20CC d) 20HS

4. CONCLUSIONS

In this study, green composite was obtained by using natural filler and inorganic filler together. The use of natural fillers in green composites provides several advantages. These include reducing environmental impact, using renewable resources, increasing recycling opportunities and promoting biodegradability. Additionally, green composite materials are generally lower cost and energy efficient.

Natural fillers also reduce the cost of the material by reducing the amount of polymer in the composite. When the amount of filler material mixed into the polymer reaches a certain value, the mechanical properties of the polymer may weaken and its hardness may increase. For this reason, the fillers used in polymer-based composites have an upper limit. In this study, the upper limit was determined as 20%. Thus, it was determined that it would reduce the cost of matrix material by 20%.

When sawdust filler is used together with calcite, it has been evaluated that the ideal ratio is 5% HS and 15% CC, however, although 10% HS and 10% CC ratios partially reduce the mechanical properties, the decrease is tolerable.

As a result of the study, it was concluded that hornbeam sawdust together with calcite can be used as a filler in polyester-based composites. This approach can contribute to reducing environmental impact in industry and preserving natural resources. With the use of green composite materials, we can move towards the goal of leaving a cleaner and more sustainable world to future generations.

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