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Investigation of the usability of laurel waste in thermoplastic composite production

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Abstract

In this study, the usability of laurel waste in thermoplastic composite production was investigated. After the laurel branch waste and leaf waste were ground and sieved, they were added to high-density polyethylene (HDPE) at a rate of 0-10-20-40% by weight and mixed in an extruder. Then, boards with dimensions of 250x250x3 mm were produced from the mixtures according to the hot press molding technique. As a result of adding branch and leaf flour to neat HDPE, tensile strength decreased. The tensile strength was determined as 22.28 MPa in the HDPE board and 8.6 MPa in the 40% leaf flour added board. As a result of adding branch and leaf flour to neat HDPE, bending strength first increased and then decreased. The highest flexural strength was determined as 30.3 MPa with 10% leaf flour additive, and the lowest bending strength was determined as 21.68 MPa with 40% leaf flour additive. According to Shore D test results, branch flour and leaf flour increased the hardness of neat HDPE. According to the thermal analysis results, it was seen that the effect of branch flour and leaf flour on the thermal properties of HDPE was limited. In addition, scanning electron microscope (SEM) images showed that HDPE and branch flour mixed better.

Keywords: Laurel, Branch and leaf waste, Thermoplastic composite, Mechanical properties

Defne atıklarının termoplastik kompozit üretiminde kullanılabilirliğinin araştırılması

Öz

Bu çalışmada, defne atıklarının termoplastik kompozit üretiminde kullanılabilirliği araştırılmıştır. Defne dal atığı ve yaprak atığı öğütülüp elendikten sonra, ağırlıkça %0-10-20-40 oranında yüksek yoğunluklu polietilen'e (YYPE) eklenerek ekstruderde karıştırıldı. Daha sonra, karışımlardan sıcak pres kalıplama tekniğine göre 250x250x3 mm ebatlarında levhalar üretildi. Saf YYPE'ye dal ve yaprak unu eklenmesi sonucunda çekme direnci azaldı. Çekme direnci YYPE levhada 22,28 MPa, %40 yaprak unu katkılı levhada 8,6 MPa olarak belirlendi. Saf YYPE'ye dal ve yaprak ununun eklenmesi sonucunda eğilme dirençleri önce arttı, sonra azaldı. En yüksek eğilme direnci %10 yaprak unu katkılısı ile 30.3 MPa, en düşük eğilme direnci %40 yaprak unu katkılısı ile 21.68 MPa olarak belirlendi. Shore D testi sonuçlarına göre dal unu ve yaprak unu saf YYPE'nin sertliğini arttırdı. Termal analiz sonuçlarına göre dal unu ve yaprak ununun YYPE'nin termal özelliklerine etkisinin sınırlı olduğu görüldü. Ayrıca taramalı elektron mikroskobu (SEM) görüntüleri YYPE ile dal ununun daha iyi karıştığını gösterdi.

Anahtar kelimeler: Defne, Dal ve yaprak atığı, Termoplastik kompozit, Mekanik özellikler

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1 Introduction

Population growth and industrialization have increased the use of petroleum-based plastics day by day, and plastic and its derivatives are used extensively in many areas such as packaging, construction and automotive. The waste produced after the use of plastic and its derivatives poses a threat to the environment and living health. The disadvantage of petroleum-based plastics and their derivatives is that they take longer to decompose in nature than natural polymers such as cellulose and starch. Polymers can be mixed with organic and inorganic fillers to reduce plastic usage and improve material properties (Klyosov 2007; Saxena et al. 2008; Kuan et al. 2021; Ramesh et al. 2022; Xu et al. 2022; Acharjee et al. 2023). Thermoplastics are the most used polymers in the production of composite materials. Thermoplastic polymers are preferred because they can be processed at low temperatures to prevent the degradation of lignocellulosic materials at high temperatures (Matuana and Heiden 2004). Thermoplastic composites are materials obtained by mixing petroleum-based polymers such as polyethylene, polypropylene and polyvinyl chloride with organic (lignocellulosic, carbon, etc.) and inorganic (silica, talc, etc.) filler/reinforcement materials and have different properties than other materials. Natural fibers used for filling or reinforcement purposes are obtained from renewable resources and are cheaper than synthetic fibers. It is known that these fibers provide good reinforcement ability to composites (Jayaraman and Bhattacharyya 2004). Wood is a material with excellent properties for many applications because it is abundant in nature, a renewable resource, and a low-cost material. (Hosseinihashemi and Arwinfar 2023). Different wood flour, annual plants and some minerals are used as fillers in the production of wood plastic composite (WPC) (Bal 2023).

Several studies have been carried out to date on the production of composites by mixing waste materials with thermoplastic polymers. Özmen et al. (2014) aimed to potentially use fiberboard wastes released during production in furniture factories instead of wood flour in the production of WPC. Çetin et al. (2014) investigated the mechanical properties of the composites that they produced by adding high-density polyethylene (HDPE) to Turkish pine (*Pinus brutia*) bark waste. Altuntaş et al. (2017) evaluated the strength properties of the composites that they obtained from a mixture of Scots pine (*Pinus sylvestris*) wood flour and HDPE. Narlıoğlu et al. (2018b) stated that polyvinylchloride (PVC) composites containing black pine wood flour and medium-density fiberboard (MDF) powder can be used in structural applications that require low mechanical properties such as wall cladding, exterior cladding, and door-window frames. Bal (2023) examined the effects of filler on the properties of composite materials produced by adding wood flour and glass flour to recycled low-density polyethylene.

Laurel, whose leaves are used as spices and whose oil is used as soap, perfume and body lotion, is also used as a food preservative thanks to its antifungal and antimicrobial properties (Yılmaz and Çiftçi 2021). The waste remaining after processing the laurel is generally disposed of by throwing it in the trash or burning it. Converting waste into high value-added products instead of burning or throwing it away is an important issue in terms of recycling and sustainability. Zakaria et al. (2022a) reported that Sentang (*Azadirachta excelsa*) leaf, branch, and stem waste improved the mechanical properties of high-density polyethylene (HDPE). In this study, it was aimed to evaluate laurel wastes in thermoplastic composite production. For this purpose, composites were produced by mixing laurel branch and leaf wastes with HDPE and the mechanical, thermal and morphological properties of the produced materials were determined.

2 Materials and Methods

2.1 Materials

Laurel wastes were provided from laurel processing factories in Hatay province, and high density polyethylene (HDPE) was provided from Petkim co. İzmir, Turkey.

2.2 Methods

2.2.1 Preparing composites

The mixed laurel wastes were classified as branches and leaves and then, ground in a laboratory type grinder. The ground branch flour and leaf flour were screened by a vibrating sieve. The flour with particle sizes between 60-80 mesh was dried at 103±2 °C. The branch flour and leaf flour 0-10-20-40% by weight was added to the neat HDPE and mixed by a twin-screw extruder set at the barrel temperature between 60-90-120-140-150-160-170-175 °C. The mixtures were compressed in a 250x250x3 mm mold at 9 tons of pressure and 180 °C for 15 minutes by a hydraulic press. Composite boards were obtained from the pellets according to the ASTM D4703. The boards were climatized at 60±5% relative humidity and 22±3 °C for one week. The formulation of the samples is given in Table 1.

Table 1. Sample formulations

Sample	Branch flour (%)	Leaf flour (%)	HDPE (%)
G1	0	0	100
G2	10	0	90
G3	20	0	80
G4	40	0	60
G5	0	10	90
G6	0	20	80
G7	0	40	60

2.2.2 Determination of mechanical properties

Tensile and flexural strength tests were performed according to ASTM D638 and ASTM D790, respectively, using an electromechanical universal testing machine. Six different samples were tested for each group in order to determine the tensile and bending properties of the boards. Shore D hardness test was performed according to ASTM D2240 to determine the hardness of the boards. Shore D tests were performed by measuring 5 different points of 6 different samples.

2.2.3 Statistical analysis

The values of mechanical test results were examined with one-way analysis of variance (ANOVA) using the SPSS Statistics 22.0 program and then subjected to the Duncan test. According to the Duncan test results, statistical differences (at 0.001 significance level) are shown in lowercase letters on the graphs where the mechanical test results are presented.

2.2.4 Determination of thermal properties

Thermo gravimetric analysis (TGA) and differential scanning calorimetry (DSC) analysis were performed to determine the thermal properties of the boards. TGA analysis was performed using the Shimadzu TGA-50 device set to 600 °C, and DSC analysis was performed using the Shimadzu DSC-60 device set to 300 °C. In thermal analysis, the heating rate was set as 10 °C/min and the nitrogen gas flow rate was set as 50 mL/min.

2.2.5 Determination of morphological properties

The cross sections of the composites were imaged using scanning electron microscopy (SEM) to examine the filler distribution in the polymer matrix and the physical relationship between the polymer matrix and the filler. The cross sections were gold coated under a high vacuum with a special device (Q150R Plus) before images were taken from the SEM device (ZEISS Sigma 300 VP).

3 Results and Discussions

3.1 Mechanical properties of samples

The tensile strengths of the test samples are shown in Figure 1. The tensile strength of the HDPE board was measured as 22.28 MPa. The tensile strength (21.12 MPa) was determined in the boards with 10% branch flour added (G1). The lowest tensile strength value (8.6 MPa) was determined in boards containing 40% leaf flour. The tensile strength of boards containing branch flour is higher than that of boards containing the same amount of leaf flour by weight, and this is attributed to the fact that the branch has a more fibrous structure than the leaf. When the results were analyzed statistically, it was seen that there was no statistically significant difference between the tensile strength value of the HDPE board and the tensile strength values of the composites containing 10% branch flour and 10% leaf flour. There was no statistical difference observed between the tensile strengths of samples with 40% branch flour and 20% leaf flour. In addition, the sample containing 20% branch flour and 40% leaf flour showed statistically different results from the other samples. In a study, it has been reported that the values of tensile strength decrease with the addition of filler in low-density polyethylene composites with wood flour and glass flour additives (Bal 2023). In another study, it was observed that the tensile strength decreased when medium-density fiberboard (MDF) flour was added to the polymer matrix (Özmen et al. 2014).

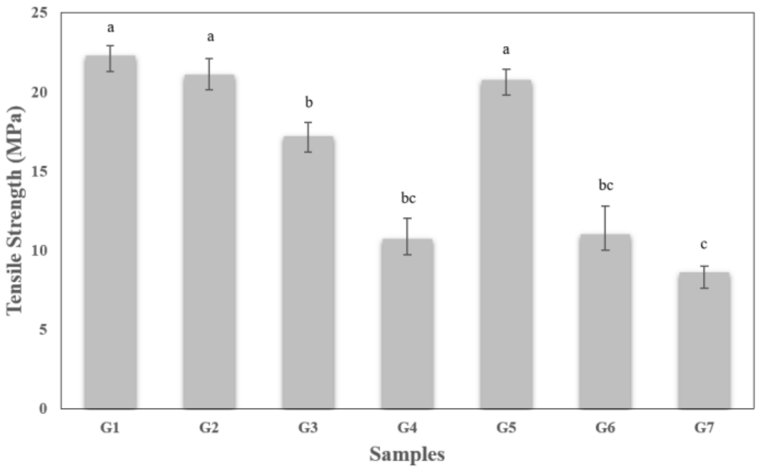

Figure 1. Tensile strengths of the samples

The modulus of elasticity of the samples is given in Figure 2. The highest tensile modulus (1599 MPa) was determined in boards containing 10% branch flour (G1). Also, the tensile modulus of samples (G5) containing 10% leaf flour was determined as 987 MPa. The lowest tensile modulus (701 MPa) was determined in the samples containing 40% leaf flour. According to the statistical analysis results, there is no significant difference between the other samples except the composite sample containing 10% branch flour. It has been stated that increasing the fiber ratio in composites produced by mixing rice straw components (rice husk, rice straw leaf, rice straw stalk and whole rice straw) and wood fiber with HDPE leads to a decrease in tensile strength and an increase in modulus (Yao et al. 2008). In this study, exhibiting different tensile strength properties depending on the sample type was attributed to the modulus of the fillers.

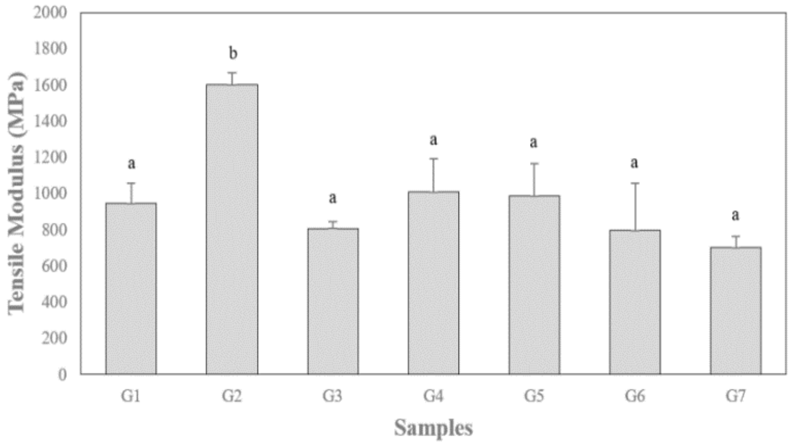

Figure 2. Tensile modulus of the samples

The flexural strength of HDPE board (G1), branch flour and leaf flour-added composites (G1-G7) are shown in Figure 3. The bending strength of the HDPE board was determined as 28.37 MPa. The flexural strengths of the samples (G2-G3-G4) containing 10-20-40% branch flour by weight were determined as 30.26 MPa, 29.42 MPa and 27.66 MPa, respectively. The highest flexural strength was determined as 30.3 MPa in the sample containing 10% leaf flour. The lowest flexural strength was determined as 21.68 MPa in the sample containing 40% leaf flour. It was seen that there was no statistically significant difference between the flexural strength of the HDPE board and the other composites except for 20-40% leaf flour-added composites. The changes in the strength of boards containing filler at the same weight ratio are attributed to the different chemical compositions of the filler (holocellulose, lignin and extractives). Lignocellulosic materials provide stress point concentrations in composites. In contrast, extractives weaken the bond with lignocellulosic fibers (Ashori and Nourbakhsh, 2010). Mu et al. (2021) reported in their study that adding agricultural waste fibers to HDPE increased the bending strength.

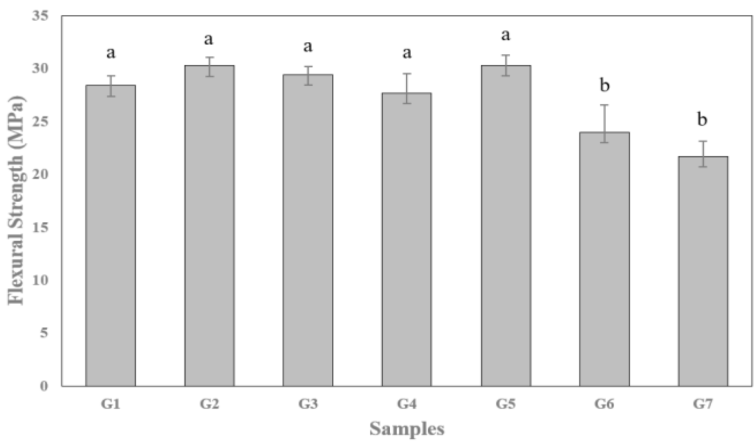

Figure 3. Flexural strengths of the samples

Figure 4 shows the flexural modulus of the samples. The flexural modulus of the HDPE board was determined as 430 MPa. The highest flexural modulus was determined as 614 MPa in the sample containing 10% branch flour. The lowest flexural modulus was determined as 382 MPa in the 40% leaf flour added sample. It has been observed that adding branch flour to the neat HDPE improves the flexural modulus compared to adding leaf flour. It was seen that there was no statistically significant difference between the 20% leaf flour added sample and the HDPE board. Also, it was observed that the samples containing 20% and 40% branch flour and the 10% leaf flour added sample were statistically similar. Bal (2022) reported that the flexural strength and modulus of elasticity increased when adding pine sawdust to linear low-density polyethylene (LLDPE). In a study, it was stated that adding black pine (*Pinus nigra*) sawdust into the polymer matrix improved the bending strength properties (Narlıoğlu et al. 2018a). Another study reported that Turkish pine (*Pinus brutia*) bark flour significantly improved the flexural strength properties of HDPE composites (Çetin et al. 2014).

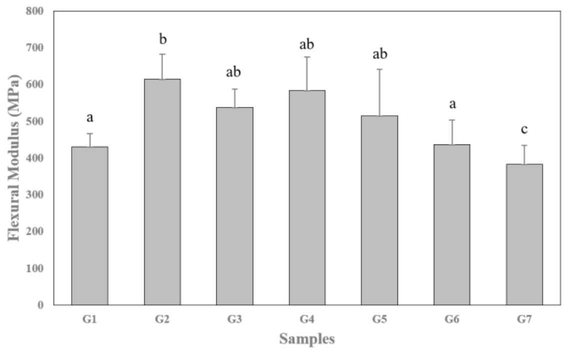

Figure 4. Flexural modulus of the samples

The Shore D hardness of the samples is given in Figure 5. The hardness of the HDPE without fillers was measured as 60.96. It has been observed that adding branch flour and leaf flour to the neat HDPE increases the hardness of the samples. The highest Shore D hardness value was determined as 63.18 in the sample containing 40% branch flour. The Shore D hardness value of the sample containing 40% leaf flour was determined as 62.28. It was seen that there was no statistical difference between the Shore D hardness of the HDPE board and the 20% leaf flour added board. Also, Shore D hardness of samples containing 20% branch flour, 10% and 40% leaf flour was found to be statistically similar. In addition, it was observed that there was no statistical difference between the hardness of boards containing 10% and 40% branch flour. In a study, the addition of wood flour and glass flour into low-density polyethylene (LDPE) increased the hardness of the composites (Bal 2023). Narlıoğlu (2021) reported that wood sanding dust waste increased the hardness of HDPE in a study evaluating hornbeam wood sanding dust waste in thermoplastic composite production.

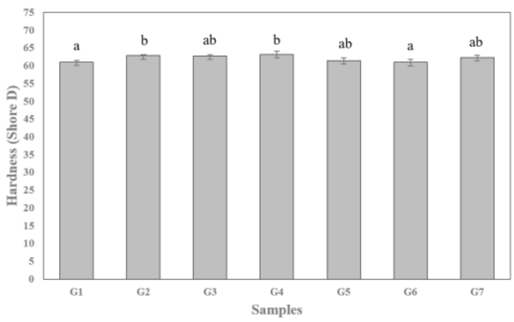

Figure 5. Shore D hardness of the samples

3.2 Morphological properties of samples

SEM images of the cross section of samples containing branch flour (G2-G3-G4) and leaf flour (G5-G6-G7) are given in Figure 6. In the cross sectional images of the branch flour added composites, it is seen that the branch flour filling and the HDPE are well mixed with each other and there are polymer extensions in the cross section. In addition, there are fewer gaps in the cross sectional images of composites containing branch flour than those containing leaf flour. Since the leaf surfaces (areas circled) seen in the cross section of leaf flour added composites are smoother, the polymer matrix adheres poorly to these surfaces.

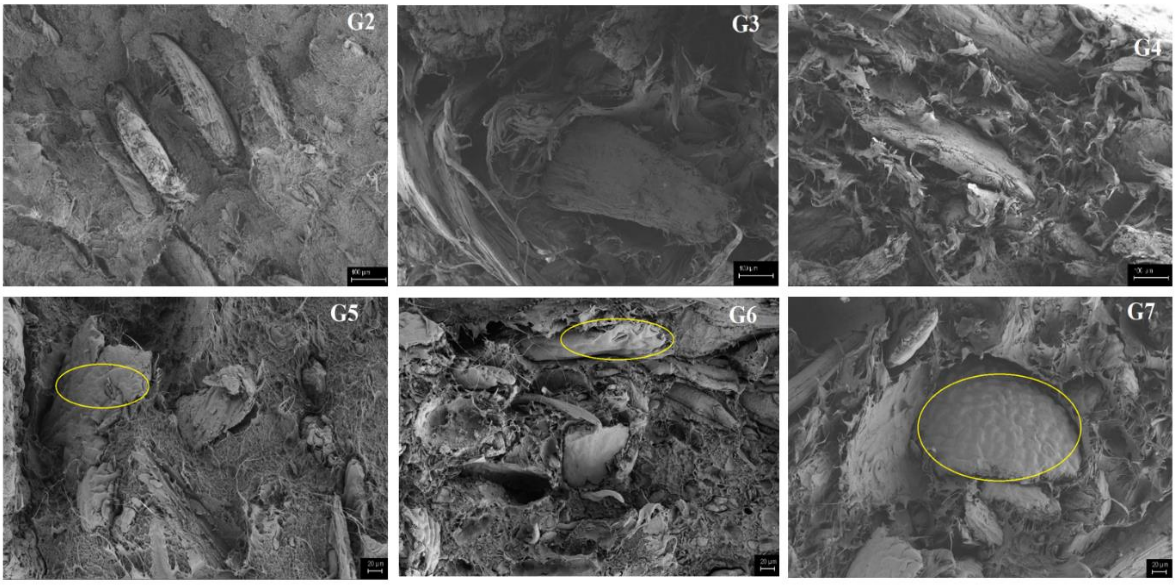

Figure 6. SEM images of samples (G2: 10% branch flour added sample, G3: 20% branch flour added sample, G4: 40% branch flour added sample, G5: 10% leaf flour added sample, G6: 20% leaf flour added sample, G7: 40% leaf flour added sample)

It can be seen that there are gaps in the cross section images (G5-G6-G7) of leaf flour added composites. It is thought that the lower mechanical properties of composites containing leaf flour compared to those containing branch flour are due to the low interfacial bond with the polymer matrix due to the leathery structure of the leaf surface. When SEM images are examined, it is seen that the cross sectional structure of the composites supports the mechanical test results.

3.3 Thermal properties of samples

TGA curves of neat HDPE, branch flour and leaf flour added HDPE composites are given in Figure 7. As can be seen from the TGA curves, the degradation curve of neat HDPE is single-stage. The average decomposition temperature of neat HDPE is at 487 °C, and the decomposition started at 472 °C and continued until 502 °C. The degradation of branch flour and leaf flour filled composites occurred in two stages. The 1st stage decomposition temperatures of branch flour added samples were determined as 337 °C, 342 °C and 331 °C for G2, G3 and G4 samples, respectively. Altuntaş and Arıkan (2022) reported that the first decomposition temperatures of composites are very close to each other and the first material to decompose is lignocellulosics. The 2nd stage decomposition temperatures of branch flour added samples were observed between 483-486 °C. The 1st stage decomposition temperatures of leaf flour added samples (G5, G6 and G7) were determined as 299 °C, 271 °C and 324 °C, respectively. It can be seen that the 1st stage decomposition temperatures of leaf flour added samples are significantly different from each other due to the chemical composition of the leaf which is less homogeneous than the branch. The 2nd stage decomposition temperatures of branch flour added samples between 487-492 °C.

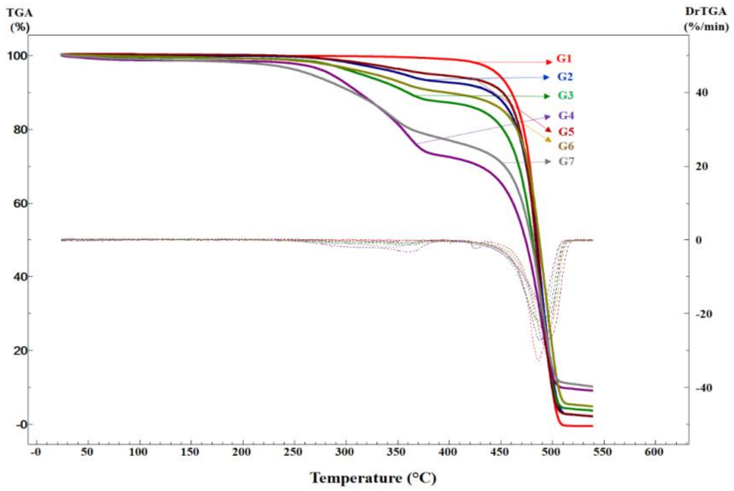

Figure 7. TGA curves of neat HDPE (G1), branch flour (G2-G3-G4) and leaf flour (G4-G5-G6) added samples

As can be seen from the TGA curves, it was observed that the addition of branch flour and leaf flour to the HDPE did not cause a significant change in the degradation temperatures. The use of wood waste particles obtained from leaves, branches and stems as fillers in the production of wood plastic composite (WPC) is preferred due to the environmental advantages and good thermal properties of the composite (Zakaria et al. 2022b).

DSC curves of neat HDPE, branch flour and leaf flour added samples are given in Figure 8. The melting temperature of neat HDPE is determined 139 °C and the melting enthalpy is 230 j/g. Adding branch flour and leaf flour to HDPE slightly changed the melting temperatures.

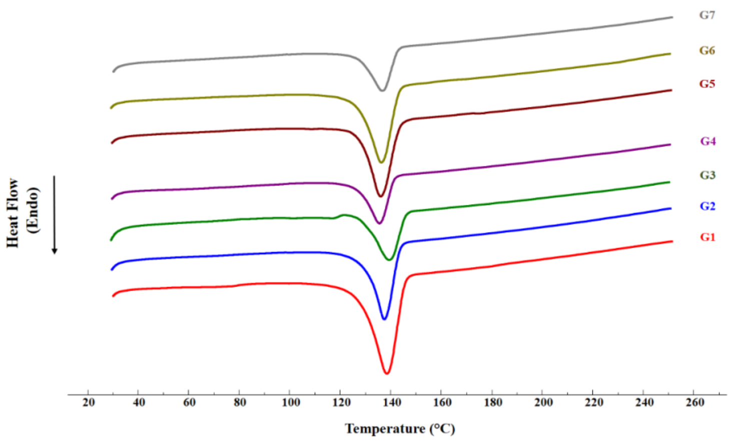

Figure 8. DSC curves of neat HDPE (G1), branch flour (G2-G3-G4) and leaf flour (G4-G5-G6) added samples

As a result of adding 10-20-40% by weight branch flour to neat HDPE, the melting temperatures were determined as 138 °C, 141 °C and 137 °C, respectively. When 10-20-40% by weight leaf flour was added to neat HDPE, the melting temperatures were determined as

136 °C, 137 °C and 137 °C, respectively. In addition, it was observed that the addition of branch flour and leaf flour to neat HDPE reduced the melting enthalpies. Similarly, Altuntaş and Arıkan (2022) reported that the melting enthalpy decreases with the increase in the amount of filler in the polymer matrix. The melting enthalpies of G2, G3 and G4 samples containing branch flour were determined as 150 j/g, 113 j/g and 97 j/g, respectively. In addition, the melting enthalpies of G5, G6 and G7 samples containing leaf flour were determined as 150 j/g, 156 j/g and 95 j/g, respectively.

4. Conclusion

In this study, boards were produced without any problems by mixing laurel branch and leaf wastes with HDPE. The results and recommendations are listed below:

- Adding branch flour and leaf flour to HDPE reduced the tensile strength. On the other hand, there was an increase in flexural strength at low filling rates.
- An increase and decrease in the elasticity modulus was observed depending on the amount of filler added to HDPE. In addition, adding branch flour and leaf flour to HDPE generally increased the hardness of the boards.
- Adding branch flour and leaf flour to HDPE did not change much in the thermal decomposition temperatures. In addition, no significant change was observed in the melting temperatures of the boards.
- In the cross sectional images of the boards examined by SEM, it was seen that the branch flour mixed well with HDPE. In addition, it was seen that there were more gaps in the cross-sectional images of leaf flour added composites and that HDPE did not mix well with leaf flour.
- The synergistic effect created by mixing branch flour and leaf flour in different ratios may be interesting. In addition, investigating the compatibility of laurel waste with different petroleum-based polymers and biodegradable polymers will contribute to the literature.

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Author Contributions

Nasır Narlıoğlu: Conceptualization (Developing research ideas and objectives), Data curation, Formal Analysis, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Hüseyin Onur Sever**: Formal Analysis, Investigation, Methodology.

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Conflict of interest statement

The authors declare no conflict of interest.

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