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Physical and mechanical properties of composites produced from recycled polypropylene and cardboard-polyethylene-aluminium mixture

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Abstract

Plastic-containing waste causes significant environmental pollution because it remains in nature for a long time without degrading. This waste includes polyolefin-based containers, polyethylene terephthalate water bottles and cardboard-polyethylene-aluminium beverage boxes. In recent years, important steps have begun to be taken to eliminate the environmental effects of solid waste. The goal is to reduce this waste by using it to produce new composite products. In this study, composite sheets were produced by mixing polypropylene (PP), obtained from recycled waste plastic chairs, as a polymer matrix and recycled waste Tetra Pak® boxes (TPBs) as filler in different mixing ratios. Density, thickness swelling, water absorption, flexural strength, flexural modulus, deformation at bending, tensile strength, tensile modulus, elongation at break and hardness were determined. According to the data obtained, it was determined that as the amount of filler in the composite increased, the density, thickness swelling, water absorption, flexural modulus, tensile modulus and hardness values increased, whereas the flexural strength, deformation at bending, tensile strength and elongation at break values decreased.

1. Introduction

In recent years, many different kinds of polypropylene-based materials, kitchenware, boxes, plastic furniture, etc. have become major environmental problems. Recycling these types of waste and obtaining new materials is important, both economically and in terms of reducing environmental waste. Additionally, liquid beverage containers containing cardboard, polyethylene and aluminium also cause significant environmental problems. Tetra Pak® boxes (TPBs) consist of 6 layers and contain 75 % paper, 20 % polyethylene and 5 % aluminium [1]. Beverage containers of this type are used on the market under the trade names Tetra Pak®, Pure-Pak®, Signature EVO® and Greatview®. Different methods have been developed to transform this type of packaging waste, including obtaining sheets by breaking down these boxes and pressing them under heat, obtaining paper fibre from these boxes using the hydropulping method, obtaining pellets (Ecoallane) from the remaining polyethylene and aluminium, and producing energy by burning these wastes [2]. Today, products such as panels, sheets, bricks and wood-polymer composites (WPCs) made from waste TPBs are used as construction materials [3].

Some laboratory studies have been conducted on the use of waste TPBs in the production of composite materials. For example, Ayrilmis et al. [1] evaluated some important properties of cardboard substrate panels overlaid with beech veneer using four different adhesives. Avella et al. [4] used maleic anhydride grafted polyethylene (MAPE) to investigate some of the properties of high-density polyethylene (HDPE)-based composite material filled with recycled TPBs. They reported that TPBs could be considered a valid alternative for common application sectors where fibre-reinforced polyolefins are already in use. Ayrilmis et al. [5] evaluated some of the mechanical properties of TPB composites without a polymer, reinforced with rice husk flour. Ebadi et al. [6,7] investigated some of the physical and mechanical properties of low-density polyethylene (LDPE)-based WPC boards filled with recycled TPBs and wood flour. They reported favourable effects when using the TPBs filler in the matrix material of WPCs. Hamouda et al. [8] investigated some of the mechanical properties and decay resistance of composite boards produced using shredded TPBs and wool yarn waste. In a similar study, Mohareb et al. [9] investigated some of the physical properties and decay resistance of composite boards produced using shredded TPBs and wool yarn waste. In another study, Aranda-García et al. [10] analysed the main processing and formulation factors that affect the performance of HDPE composites filled with recycled TPBs. They reported that the processing time plays a fundamental role in the mechanical properties of HDPE composites filled with recycled TPBs. In a different study, Hidalgo [11] investigated the effects of low and high pressure on selected properties of TPB composites and reported that waste TPBs are a useful recycling resource for conversion into rigid boards.

Bekhta et al. [12] evaluated some of the properties of experimentally obtained composite panels manufactured from waste TPBs, food packaging films and candy polyethylene wrappers without using any additional binders and without using an extruder. They reported that this waste material may have the potential to be used as raw material for the production of new composites without the use of adhesives in the panels. Platnieks et al. [13] investigated the use of recycled cellulose separated from TPBs in an industrial processing plant as a structural filler for biocomposites produced from polybutylene succinate and reported some favourable biological, physical and mechanical test results. Sanchez-Cadena et al. [14] evaluated the mechanical properties and fracture behaviour of composites produced with recycled HDPE and TPBs without using an extruder. In a different study, Sujatha et al. [15] investigated some of the mechanical properties of composite boards produced from recycled TPBs with and without resin and reported some favourable results for TPB-based composite boards with resin. Üner and Bülbül [16] investigated some of the mechanical properties of composite boards produced from recycled TPBs and polyethylene (PE) using some fillers and binding agents. Auriga et al. [17] investigated some of the physical and mechanical properties of chipboard produced from recycled TPBs using urea-formaldehyde resin. Bal [18] compared the mechanical properties of PE-based composite sheets filled with recycled TPBs and wood flour and some favourable results were reported according to the data obtained.

In some of these previous studies, the waste TPB material was first cut into small pieces, mixed with different polymers and pressed in a hot press to produce composite sheets. In other studies, the waste TPB material was first cut into small pieces and then glue was added and composite sheets were produced after hot pressing. In others, TPBs were first shredded, ground with a grinder and mixed in an extruder before composite sheets were produced by being moulded in a hot press. In summary, there were a limited number of previous studies that ground TPBs and mixed them in an extruder. Moreover, to the best of the author's knowledge, no previous study used waste polypropylene (PP) as a polymer and waste TPBs as a filler. This study aimed to investigate the effect of the filler amount on some of the properties of a composite material produced using waste PP as a polymer matrix and ground waste TPBs as a filler.

2. Experimental details

2.1 Materials

In this study, PP-based waste plastic chairs were used as a polymer matrix. The waste plastic chairs (recycled polypropylene, denoted as R-PP) were first cut into small pieces (Fig. 1a) and then these pieces were ground in a high-speed grinder (Demsan Brader 1500) to make them smaller (Fig. 1b). The cardboard-polyethylene-aluminium boxes used in the study were Tetra Pak brand milk cartons (recycled Tetra Pak Box, denoted as TPB). TPB is a multi-layered packaging type designed to keep food and beverages fresh. It is composed of three materials, i.e. paper (70 %), polyethylene (24 %) and aluminium (6 %). These TPBs were collected by the authors from their household waste. The insides were washed and dried. Then, they were cut into small pieces (Fig. 1c) and ground in a high-speed grinder (Demsan Brader 1500) to obtain a wool-like material (Fig. 1d). Every 100 grams of TPBs were placed into the grinder, the lid was closed and grinding was done for about 1 minute.

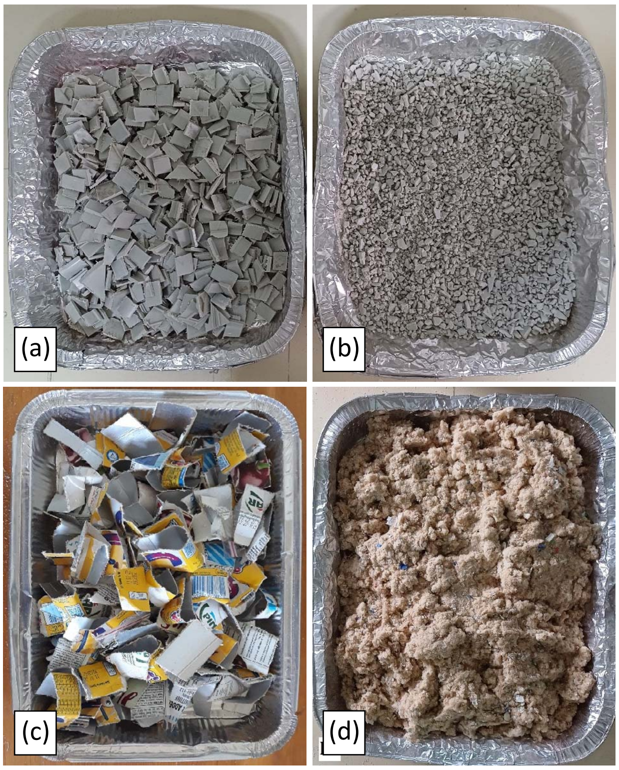

Figure 1. Materials: (a) waste plastic chairs pieces, (b) ground waste plastic chairs pieces, (c) shredded TPBs and (d) ground TPBs

2.2 Preparation of composites

The composition of the composites is given in Table 1. The R-PP and TPB materials were dried at 103 ± 2 °C and then mixed to obtain a homogenous blend before processing in a single screw extruder (Fig. 2a). The temperature of the extruder barrel was adjusted to 150, 160 and 175 °C degrees from the feeding section to the exit section, respectively. The screw speed was adjusted to 30 rpm. The extruded blend was taken in filament form from the barrel exit, which had a nozzle diameter of 3 mm. The filaments were cooled in the air on a desk (Fig. 2b). The cooled blend was cut into pellets and these pellets were ground using a grinder. The ground blend was remixed with the extruder. The filaments were again cooled in the air on a desk and cut into pellets. These pellets were placed in a metal mould and transferred to a hot press at a temperature of 175 ± 5 °C. A lubricant agent (WD-40 multi-use product) was applied to the metal surface to prevent the pellets from sticking to the mould. The blend was heated and melted over a period of 15 minutes. At the end of this period, the metal mould containing the mixture was removed from the heater and immediately placed in a cold press. A total of 16 MPa of pressure was applied in the cold press for 5 minutes. The plate was taken from the metal mould and a composite sheet was thus obtained with the dimensions (thickness × width × length) of 3.5 × 175 × 175 mm. Four composite sheets were produced for each group, with a total of 20 sheets for the whole study (Fig. 2c). Test samples were prepared from these sheets. A total of sixteen test samples were prepared for each test by cutting four test samples from each sheet. The test samples were cut using a laboratory band saw. The edges of each test sample prepared for the tensile test were shaped with a CNC machine.

Table 1. Composition of composites

Group	1	2	3	4	5
R-PP, wt. %	100	90	75	60	40
TPB, wt. %	0	10	25	40	60

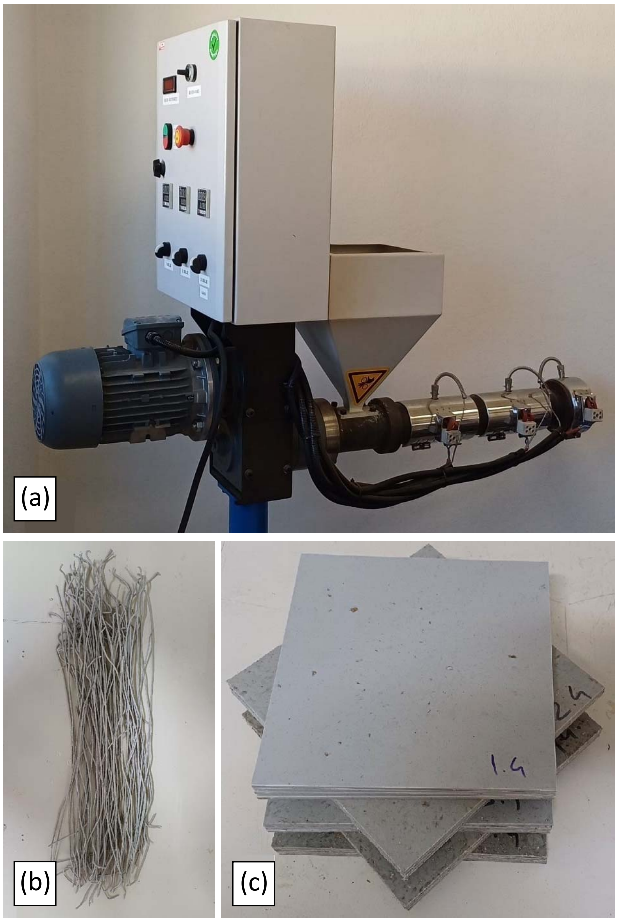

Figure 2. Preparation of composites: (a) single screw extruder, (b) filaments and (c) composite sheets

2.3 Characterisation of composites

The density of each composite material was determined based on ASTM D792 [19]. The density values were measured by dividing the weight of each test sample in air by its volume in water. Measurements were made on 16 test samples for each group.

Flexural tests were conducted according to ASTM D790 [20] using the electromechanical universal testing machine (Natek), as can be seen in Figure 3a. Flexural test samples were prepared with the dimensions (thickness × width × length) of 3.5 × 20 × 80 mm. Measurements were made on 16 test samples for each group to determine the flexural strength. In the flexural strength test, the preload was set to 2 N, the support opening was 56 mm, the test speed was 2 mm/min and the test ended when the specimen reached 75 % of the maximum measured force. The maximum deformation values at the end of the flexural tests are given in the tables as the deformation at bending.

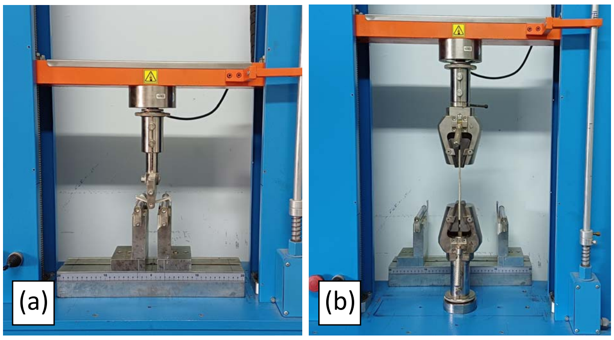

Figure 3. Universal testing machine: (a) flexural test and (b) tensile test

Tensile tests (Fig. 4b) were conducted according to ASTM D638 [21]. A tensile test was performed on test samples formed in a "dog-bone" shape described in ASTM D638. For the tensile test, 16 test samples from each group were prepared. The tensile test samples were prepared with the dimensions of 3.5 × 20 × 165 mm. The middle area of each tensile test specimen was shaped with a CNC machine. The preload was set to 5 N and the test speed was set to 2 mm/min. The distance between grips was 115 mm. At the end of the tensile test, the maximum percentage of elongation at break was also calculated. This value is given in the tables as the elongation at break.

The Shore D value was determined according to ASTM D2240 [22]. Hardness tests were performed on the Shore D test device (Loyka LXD-D). For the hardness test, 16 test samples from each group were prepared.

Test samples measuring 3.5 × 20 × 20 mm were used to determine thickness swelling and water absorption according to the ASTM D570 [23]. The test samples were immersed in water for 24 hours. At the end of the immersion time, they were removed from the water and the surface was cleaned with a dry cloth. The specimens were weighed and measured to calculate thickness swelling and water absorption.

The elastic modulus values measured during the bending and tensile tests are also given in the tables. One-way ANOVA and Duncan tests [24] were performed on the data obtained at the end of the tests using a statistical program (SPSS 13.0) and the results are shown in the relevant tables. Applying the Duncan multiple comparison test at $\alpha = 0.05$, arithmetic means followed by the same letter are not significantly different from each other.

3. Results and discussion

Table 2 lists the density data for the test samples from the control group without filler (group 1) and other experimental groups with filler (groups 2, 3, 4 and 5), along with the Duncan test results and p-values showing the ANOVA test results for these data. The lowest density was found in group 1 and the highest density was found in group 5 with values of 988 and 1107 kg/m^3, respectively. No filler was added to group 1 whereas 60 wt. % TPB filler was added to group 5. The density value of the composite material increased with the addition of the filler. A statistically significant (p-value < 0.001) difference was found between the density values of all the groups. The measured density of the recycled PP used in the study was 988 kg/m^3. The density of virgin PP generally varies between 855 and 920 kg/m^3. The recycled PP used in the study was obtained from waste plastic chairs. Fillers such as calcite added to the PP material cause the density of this polymer to increase during chairs production. Therefore, the measured density of the control group was slightly higher than that of virgin PP. Likewise, the measured densities of other experimental groups were higher than that of the control group because the TPB filler added in this study increased their densities. Similar results have been reported in previous studies using TPB as a filler [7,10,25]. In previous studies on polymer composite materials, it was determined that the density of the new composite material obtained increased as a result of the filler's high density [16,18,26-28].

Table 2. Density test data, ANOVA p-value and Duncan test results

Group	1	2	3	4	5	p-value
$\bar{X}$, kg/m^3	988	1020	1047	1078	1107	0.000
SD, kg/m^3	6.5	5.1	9.8	7.5	11.5	
Duncan test	A	B	C	D	E	

$\bar{X}$ – arithmetic mean; *SD* – standard deviation

The water absorption and thickness swelling test results obtained at the end of the tests are given in Table 3. It can be seen that the lowest water absorption percentages and thickness swelling percentages were obtained in group 2 and the highest were found in group 5. In general, the water absorption and swelling percentages increased with the amount of TPB filler. The differences between groups were statistically significant (p-value < 0.001). According to the Duncan test results, the water absorption and thickness swelling percentages of all the experimental groups differed. In this study, the most important factor affecting the water absorption and thickness swelling percentages was the hydrophilic wood fibres that made up the cardboard in the TPB material. The cellulose and hemicelluloses contained in these wood fibres are highly hydrophilic and therefore absorb water. Similar results were obtained by other researchers as well [7,10,12,25].

Table 3. Thickness swelling and water absorption test data, ANOVA p-values and Duncan test results

Group	1	2	3	4	5	p-value
Thickness swelling						
$\bar{X}$, %	–	0.89	1.60	2.78	4.11	0.000
SD, %	–	0.22	0.63	0.66	0.65	
Duncan test	–	A	B	C	D	
Water absorption						
$\bar{X}$, %	–	0.72	1.33	2.71	4.64	0.000
SD, %	–	0.26	0.30	0.47	0.44	
Duncan test	–	A	B	C	D	

$\bar{X}$ – arithmetic mean; *SD* – standard deviation

Table 4 lists the data obtained in the bending tests, ANOVA test significance levels and Duncan test results. According to these data, the highest bending strength was obtained in group 1 without filler (36.9 N/mm^2) and the lowest bending strength was obtained in group 5 (26.3 N/mm^2) which contained 60 wt. % TPB filler. As the amount of filler increased, the bending strength decreased. The differences between the groups were statistically significant (p-value < 0.001). In previous studies, as the ratio of particulate or fibrous filler used in PE-based or PP-based composite materials increased, the bending strength of the composite material decreased [16,29,30]. It has been reported that one of the most important reasons for this situation to occur is the lack of sufficient interfacial adhesion of composites produced from polar and nonpolar materials [28,29]. In addition, the incompatibility of the hygroscopic filler with the hydrophobic polymer causes the bending strength of the composite material to decrease. In this study, the TPB used as a filler consisted of three different materials (cardboard, aluminium and polyethylene). These had very different properties which negatively affected the bending resistance of the composite. In this regard, Ebadi et al. [7] showed that as the amount of wood flour filler decreased and the amount of TPB filler increased, the bending strength of the composite increased. In another study, Bal [18] determined that the bending strength of a recycled PE-based composite material filled with wood flour and TPB increased with the TBP amount. However, the bending strength of a composite filled with 40 wt. % TPB material was similar to that of the control group. In a study conducted by Aranda-García et al. [10], the bending strength of a TPB-HDPE composite material decreased as the TPB amount increased.

Table 4. Flexural test data, ANOVA p-values and Duncan test results

Group	1	2	3	4	5	p-value
Flexural strength						
$\bar{X}$, N/mm^2	36.9	34.7	31.2	28.4	26.3	0.000
SD, N/mm^2	2.5	2.1	2.3	2.1	4.2	
Duncan test	E	D	C	B	A	
Flexural modulus						
$\bar{X}$, N/mm^2	1392	1504	1565	1639	1705	0.000
SD, N/mm^2	72	87	85	74	57	
Duncan test	A	B	C	D	E	
Deformation at bending						
$\bar{X}$, mm	19.9	9.7	7.5	6.1	5.0	0.000
SD, mm	2.1	1.1	0.9	0.7	1.0	
Duncan test	E	D	C	B	A	

$\bar{X}$ – arithmetic mean; *SD* – standard deviation

In a study conducted by Bekhta et al. [12], the bending strength of a composite material produced with TPB and polyethylene food packaging films decreased as the TPB amount increased. However, in that study, the mixture was not processed in an extruder.

The deformation at bending values given in Table 4 show that the obtained data, ANOVA significance level and Duncan test results are similar to the bending strength results. Similar deformation at bending data were obtained in previous studies conducted with thermoplastic-based polymers [27,31-33].

Flexural modulus data are given in Table 4. These data show that the flexural modulus increased with the amount of filler. The differences between the groups were statistically significant (p-value < 0.001). According to the Duncan test results, the flexural modulus values of all the groups differed. In this case, the flexural strength and flexural modulus were affected in different ways by increasing the filler material. As a general rule, flexural strength is the ability of a composite material to stay together when bent. The flexural modulus is a sign of how well the same composite material resists bending. This situation can also be explained by the general composite rule. The properties of a composite material are similar to the properties of its components. In this case, the flexural modulus of the composite material increased with the density of the composite. Similar results have been obtained by other researchers as well [7,18,29,34,35].

The tensile test data, ANOVA significance levels and Duncan test results are given in Table 5. It was determined that as the filler material increased, the tensile strength and elongation at break decreased and the tensile modulus increased. The highest tensile strength and elongation at break were measured in group 1 and the lowest tensile strength and elongation at break were measured in group 5. In contrast, the largest tensile modulus was measured in group 5 and the lowest tensile modulus was measured in group 1. The differences between the groups were statistically significant (p-value < 0.001). However, there was no difference between group 1 and group 2 according to the Duncan test. The PP used in this study was obtained from waste chairs and was recycled PP. Therefore, the tensile and bending test results of the control group test samples were expected to be lower than those of virgin PP. This occurred because some mineral substances such as calcite are added to the mixture during the production of chairs with PP material. Similar results were obtained by other researchers as well. Bal et al. [33] obtained a value of 17.5 N/mm^2 for the tensile strength of a recycled PE-based composite material produced with PP obtained from waste plastic chairs and without using fillers, while the elongation at break was 4.7 %. The tensile strength decreased with the use of wood flour filler. Similar results were obtained for composite materials produced with polyolefin group polymers. In general, as the amount of fillers added to the polymer material increases, the tensile strength and elongation at break of the composite material decrease and the tensile modulus increases [18,25-28,32-36].

Table 5. Tensile test data, ANOVA p-values and Duncan test results

Group	1	2	3	4	5	p-value
Tensile strength						
$\bar{X}$, N/mm^2	16.4	13.8	11.5	10.1	9.2	0.000
SD, N/mm^2	0.6	1.4	1.3	1.1	1.5	
Duncan test	E	D	C	B	A	
Tensile modulus						
$\bar{X}$, N/mm^2	446	480	695	871	872	0.000
SD, N/mm^2	49	61	110	210	159	
Duncan test	A	A	B	C	C	
Elongation at break						
$\bar{X}$, %	4.8	3.3	2.7	2.2	2.1	0.000
SD, mm	0.7	0.6	0.7	0.8	0.5	
Duncan test	D	C	B	A	A	

$\bar{X}$ – arithmetic mean; *SD* – standard deviation

The Shore D hardness test data, ANOVA test significance levels and Duncan test results for the composite materials are given in Table 6. It can be seen that the hardness values of other experimental groups were higher than that of the control group. The hardness values obtained ranged between 71 and 73. The difference between the groups was statistically significant (p-value < 0.001). It was determined that the hardness increased with the amount of filler in the composite. However, the differences between groups 3, 4 and 5 were insignificant according to the Duncan test results. Similar results were obtained by other researchers as well. The hardness value of the composite material increased as the amount of TPB decreased and the amount of wood flour increased in a study conducted by Bal [18]. The TPB amount had less effect than the wood flour on the hardness of the composite material. Additionally, previous studies on wood-plastic composite materials have reported that the hardness value of the composite material increased with the amount of filler [26,27].

Table 6. Hardness (Shore D) test data, ANOVA p-value and Duncan test results

Group	1	2	3	4	5	p-value
$\bar{X}$	71.28	72.58	73.31	73.45	73.52	0.000
SD	0.32	0.49	0.42	0.34	0.46	
Duncan test	A	B	C	C	C	

$\bar{X}$ – arithmetic mean; *SD* – standard deviation

4. Conclusion

In this study, a composite material was produced using waste plastic chairs as a polymer matrix and waste TPB material as a filler, and some of its physical and mechanical properties were determined. Based on the data obtained, the following conclusions can be drawn.

The density, thickness swelling and water absorption of the composite material increased with the amount of filler (TPB). As the amount of filler in the composite sheets increased, the flexural bending and deformation at bending decreased but the flexural modulus increased. As the amount of filler increased, the tensile strength and elongation at break decreased but the tensile modulus increased. The hardness of the composite material increased with the addition of filler compared to the control group. However, this increase was not linear.

When all the obtained physical and mechanical properties are evaluated, it can be said that TPB material can be recycled by using it in the production of composite materials as seen in this study. The produced composite material can be used as a covering material in the construction industry.

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