



Article Recycling of Nonwoven Waste Resulting from the Manufacturing Process of Hemp Fiber-Reinforced Recycled Polypropylene Composites for Upholstered Furniture Products

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Abstract: Waste recycling is a solution that reduces the environmental impact of waste landfilling or incineration. The aim of this paper is to investigate both the effect of incorporating recycled fibers obtained by defibrating 50/50 hemp/rPP nonwoven waste and the effect of the compatibilizer on the properties of composite materials. Composites incorporating 50% and 100% recycled fibers were treated with 2.5% and 5% maleated polypropylene (MAPP), respectively, and compared to both the untreated composites and the composite obtained by thermoforming from the nonwovens that generated the waste. The incorporation of 50% and 100% recycled fibers into composites decreased the tensile strength by 17.1–22.6%, the elongation at break by 12.4–20.1%, the flexural strength by 6.6–9%, and flexural modulus by 10.3–37%. The addition of 5% MAPP showed the greatest improvements in mechanical properties of composites containing 100% recycled fibers, as follows: 19.2% increase in tensile strength, 3.8% increase in flexural strength, and 14.8% increase in flexural modulus. Thermal analysis established that at temperatures ranging between 20 °C and 120 °C, the composites were thermally stable. SEM analysis revealed good coverage of the reinforcing fibers, and EDX analysis confirmed the presence of the compatibilizing agent in the structure of the composite material.

Keywords: nonwoven waste; recycling; recycled polypropylene; hemp reinforcement; composite material; compatibilization; MAPP

1. Introduction

In recent years, the usage of composite materials in the furniture industry has evolved continuously due to their benefits over traditional materials such as wood, metal, and plastics. Composite materials are very attractive for the furniture industry because they offer great flexibility in the geometrical and aesthetical design of three-dimensionally shaped products [1–4]. The combination of different possible reinforcements and matrices provides composite materials with a wide range of properties that are suitable for their end use (chairs, stools, tables, shelves, cupboards, sofa frames, racks, etc.). Composite materials for furniture applications have been obtained using different reinforcement fibers, both natural and synthetic, such as carbon [5], glass [6], hemp [7–9], flax [10], jute [11], kenaf [12], coir [13–15], oil palm [16,17], banana [18], and wood [19–21]. Nowadays, carbon and fiberglass reinforcements are increasingly being replaced by natural fibers that make composite materials environmentally friendly [22–25], sustainable [26–29], lightweight [30–32], and affordable [33–36], in addition to providing good mechanical properties [37–40].

Composite materials that contain 50% hemp fibers as reinforcement and 50% recycled polypropylene (rPP) as a matrix were produced by thermoforming by a Romanian furniture



Citation: Ichim, M.; Filip, I.; Stelea, L.; Lisa, G.; Muresan, E.I. Recycling of Nonwoven Waste Resulting from the Manufacturing Process of Hemp Fiber-Reinforced Recycled Polypropylene Composites for Upholstered Furniture Products. *Sustainability* **2023**, *15*, 3635. https://doi.org/10.3390/su15043635

Academic Editors: Valentina Siracusa, Nadia Lotti, Michelina Soccio and Alexey Iordanskii

Received: 2 February 2023 Revised: 12 February 2023 Accepted: 13 February 2023 Published: 16 February 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). manufacturing company. The selected materials allow for the manufacturing of goodquality sustainable products for a low price. Traditionally, the structural frame of an upholstered furniture product is made of wood. Due to the environmental and economic benefits, wood is being increasingly replaced by composite material. Compared to wood, which has a long production cycle, hemp plants, which provide the reinforcing fibers for the composite material, are harvested annually. Moreover, the hemp production per hectare is higher than the average yearly wood production. Therefore, by replacing wood with composite materials in the furniture industry, the human pressure on forests can be reduced, with significant environmental benefits concerning climate regulation and pollution prevention [41].

Hemp fibers are lignocellulosic fibers extracted from the stem of the Cannabis Sativa plant through stem retting, drying, and crushing, followed by scutching and hackling of fiber bundles [42]. Hemp is an environmentally friendly renewable resource that is easily available, cheap, and biodegradable and characterized by good mechanical properties and low density (compared to synthetic fibers) [43,44]. The properties of hemp fibers confer a considerable potential for their use as a reinforcement agent in polymer matrix composite materials. The waste consisting of short hemp fibers removed in the scutching/hackling process represents a valuable source of raw material that can be used in composite material manufacturing [45,46].

Polypropylene (PP) is a recyclable thermoplastic polymer with a share of 19.4% in the demand of European plastics converters in 2019 [47]. Due to its properties, such as low density, low price, good processability, good corrosion, and impact and abrasion resistance, polypropylene is used in many applications in packaging, the textile and automotive industries, building and construction, agriculture, and the electrical and electronics industries [47]. Taking into account that polypropylene is widely used in the packaging of products with a short lifespan, a large amount of polypropylene waste is generated after the product's life cycle has ended. A part of this waste is recycled, but the major part is either discarded in landfills or incinerated for energy recovery purposes. This waste disposal method results in environmental pollution, with harmful consequences for humans and biosystems. The increase in PP waste recycling reduces the environmental impact, brings economic benefits, and ensures a circular economy. In order to obtain rPP fibers, the PP waste goes through the following steps: collection, sorting, shredding, washing, melting and extrusion into pellets/flakes/granules, remelting, and extrusion into fibers [48–50]. Numerous researchers have studied the structure and properties of rPP fibers and their use in several applications, either as matrix in composites or as reinforcement in concrete [51–61].

The problem associated with natural fiber-reinforced polypropylene composites is a low compatibility between hydrophilic fibers and the hydrophobic matrix, which leads to poor adhesion at the matrix–fiber interface and therefore to poor mechanical properties of the composite materials. In order to overcome this disadvantage, the properties of the reinforcing fibers can be modified by physical modification techniques, such as corona, plasma, and alkali treatments, or by chemical modification techniques, such as esterification-based treatments (acetylation, propionylation, or benzylation), graft polymerization, use of silane coupling agents, and treatment using isocyanates [62,63]. The polypropylene matrix can also be modified by grafting maleic anhydride (MAPP), glycidyl methacrylate, and trimethylolpropane triacrylate or by surface peroxidation with heterofunctional polyperoxides [64–70].

The manufacturing process of hemp fiber-reinforced recycled polypropylene composites generates waste in the form of nonwoven fabric scraps. This category of waste can be recycled and used within the same manufacturing process in the furniture industry or in other industries for the production of various products. The possibility of using different forms of textile waste in the production process of polymeric composites process has been investigated by several authors [71–78]. The results of these studies showed that composites that contain textile waste can be used in several applications, such as the automotive, building and construction, and furniture industries.

The aim of this research was to investigate the possibility of recycling hemp/rPP nonwoven waste into thermoformed composites for upholstered furniture products. The 50/50 hemp/rPP nonwoven waste resulted from the cutting process of nonwoven fabrics according to the pattern outline in the fabrication process of 3D composites for furniture applications. The nonwoven waste was defibrated, and the resulting recycled fibers were reintroduced in new nonwovens using reincorporation rates of 50% and 100%. The novelty of this work lies in the recycling of nonwoven waste within the same manufacturing process that generated it. Thus, the length and thickness of the reinforcing hemp fibers could be examined before and after the nonwoven recycling process. Since the length of recycled fibers is shorter than the length of original fibers, it is expected that the mechanical properties of the composite materials obtained by thermoforming of nonwovens that incorporate recycled fibers would deteriorate. To compensate for this supposed depreciation of the composite mechanical properties, an MAPP compatibilizing agent was used to improve the interaction between the recycled reinforcement fibers and the matrix. The obtained composites were evaluated in terms of mechanical and thermal properties (TG, DTG, and DSC), surface morphology, and chemical analysis (SEM and EDX).

2. Materials and Methods

2.1. Materials

In the composite materials, recycled polypropylene was used as the matrix, and hemp fibers were used as reinforcement. The rPP fibers were supplied by TAPARO Company (Târgu Lăpuș, Romania) and had the following characteristics: 9.9 dtex linear density, 25.2 cN/tex tenacity, 142.9% breaking elongation, 82.2 mm mean length, and 8.2 g/10 min melt flow index (at 230 °C/2.16 kg). Hemp fibers of 15.8 tex linear density and 38.9 cN/tex tenacity were purchased from Hempflax Europe (Pianu de Jos, Romania) and were cut to 60 mm using a cutting machine.

Recycled fibers resulting from the defibration of 50/50 hemp/rPP nonwoven scraps were reincorporated into new nonwovens from which composites were obtained. These fibers differed from the reference fibers in length, as the defibration of the nonwovens resulted in fiber shortening. Nonwoven defibration may also affect the fineness of hemp fibers because of the separation of the bundles of elementary fibers from the technical hemp fibers.

Since the characteristics of the reinforcing fibers influence the mechanical properties of the composites, the length of both raw and recycled hemp fibers was measured. Single-fiber length measurement was the technique used to determine the length distribution of hemp fibers tested before and after recycling. The fibers were stretched using tweezers along a millimeter scale. Two hundred measurements were performed for each sample. The thickness of raw and recycled hemp fibers was measured using a Mesdan Analyzer 2000 microscope (Puegnago sul Garda, Italy). Fifty fibers were analyzed. Because of variations in thickness along the fiber, multiple measurements were averaged for each fiber.

Maleic anhydride-grafted PP Polybond 3200 from Crompton Corporation (Middlebury, CT, USA) supplied by S.C Prociv SRL (Bucharest, Romania) was used to improve the compatibility between the matrix and the reinforcing fibers.

2.2. Manufacturing of the Composite Materials

The Romanian furniture manufacturing company uses thermoforming to produce 3D furniture pieces from composite materials. Thermoforming is a composite manufacturing technique that allows for a high content of reinforcing fibers and a high production rate [79]. The production process consists of the following steps: manufacturing of 50/50 hemp/rPP nonwoven fabrics, overlapping and cutting of nonwoven fabrics as per the shape of the pattern, heating of fibrous mat pieces in a hot press, transferring of the heated material to a mold, pressing, and cooling. The cutting step generates up to 20% waste in the form of nonwoven scraps that can be recycled into the same product in the same manufacturing process, making the overall process more sustainable and greener.

In the experiments, the nonwoven waste was cut on a cutting machine and defibrated using an opening machine. The recycled fibers consisting of 50% hemp and 50% rPP were reintroduced in new nonwovens proportions of 50% and 100%, respectively. The manufacturing process of nonwoven fabrics consisted of fiber opening and blending, aerodynamic web formation, and mechanical web consolidation by needle punching. Six layers of rectangular nonwovens were pressed between the heating plates of the thermoforming machine in order to obtain composite plates. When laying the nonwovens, the layers were alternated according to the direction of punching (longitudinal and transversal). The thermoforming parameters were set as follows:

- Temperature: 190 °C;
- Pressure: 0.735 MPa;
- Pressing time: 15 min;
- Cooling time: 10 min.

Since defibration of nonwoven waste affects the length of recycled fibers, a decrease in the tensile strength of the composites that contain such fibers is expected. In order to counteract the effect of recycled fiber shortening on the mechanical properties of the composites, 2.5% wt and 5% wt of MAPP compatibilizer was used. The granules were ground with a ball mill and sieved between layers 2–3 and 4–5 of the nonwoven stack (Figure 1).



Figure 1. Flow chart of the composite manufacturing process.

Table 1 shows the composition and the coding of the manufactured composite variants.

Table 1.	Experimental	variants.
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Variant Code	Composition
V1	50% hemp/50% rPP
V2	100% recycled fibers from V1 nonwoven waste
V2.1	97.5% V1 recycled fibers/2.5% MAPP
V2.2	95% V1 recycled fibers/5% MAPP
V3	50% V1 recycled fibers/25% hemp/25% rPP
V3.1	47.5% V1 recycled fibers/25% hemp/25% rPP/2.5% MAPP
V3.2	45% V1 recycled fibers/25% hemp/25% rPP/5% MAPP

2.3. Mechanical Properties

Both flexural and tensile tests of composite materials were carried out on an LBG testing machine (Azzano San Paolo, Italy). The length of the specimens for tensile testing was 250 mm, and the width was 25 mm, according to the EN 326-1 standard. Tensile

test parameters were established according to ISO 527-4 standard as follows: 2 mm/min crosshead speed and 150 mm distance between clamps. The dimensions of the specimens for the 3-point flexural test were determined according to the specifications of the ISO 14125 standard. The specimen width was 15 mm, and the length was set depending on the specimen thickness. The cross-head speed used during the flexural tests was 2 mm/min. Five specimens were tested to determine the tensile and flexural properties of the composite materials.

2.4. Thermal Properties

The thermal degradation of rPP, hemp fibers (H), and hemp/rPP composite materials was studied by thermogravimetric analysis (TGA) using a Mettler Toledo TGA/SDTA 851 balance (Columbus, OH, USA). The weight of the samples ranged between 2 and 5 mg. The samples were subjected to heating in the temperature domain from 25 °C to 700 °C under a constant flow of nitrogen of 20 mL/min using a heating rate of 10 °C/min.

Differential scanning calorimetry (DSC) was used to determine the melting and crystallization temperatures of rPP fibers and hemp/rPP composite materials. The analysis was performed on a Mettler-Toledo DSC1 822e calorimeter (Columbus, OH, USA). The samples, weighing approximately 5 mg each, were placed in 40 μ L aluminum crucibles and were scanned from -60 °C to 200 °C. The first heating stage was followed by cooling and then by a second heating stage in the same temperature range. All three thermal stages were conducted at a constant rate (10 K/min) under a constant nitrogen flow rate (150 mL/min). The heating–cooling–heating cycle of the samples allows for the determination of the crystallization and melting temperature and crystallization and melting enthalpy. The crystallinity of the composite materials (X_C(%)) was calculated using Equation (1) [80]:

$$X_{c}(\%) = \frac{\Delta H_{f}}{\Delta H_{f}^{0}} \cdot 100, \tag{1}$$

where ΔH_f is the enthalpy of fusion per unit mass of the PP calculated based on the area under the melting peak of the composite, and ΔH_f^0 is the enthalpy per unit mass of the 100% crystalline PP, with a value of 207 J/g [80].

2.5. Surface Morphology and Chemical Analysis (SEM and EDX)

The surface morphology of composite materials was analyzed through a Quanta 200 (FEI) scanning electron microscope (SEM) (Hillsboro, OR, USA) operating at an accelerating voltage of 20 kV with secondary electrons in low vacuum mode. The microscope was coupled with an energy-dispersive X-ray (EDX) system for chemical analysis and elemental analysis mapping.

3. Results and Discussions

3.1. Fiber Characteristics

Figure 2 presents the fiber length distribution of hemp fibers tested before and after recycling.

It can be seen that the length distribution of hemp fibers changed considerably after nonwoven waste recycling. The average length of hemp fibers decreased from 57.8 ± 36.7 mm (before recycling) to 24.8 ± 13.9 mm (after recycling) due to the mechanical actions to which the fibers were subjected during waste cutting and opening. This represents a significant decrease (-57.09%) in hemp fiber length after recycling. In the raw hemp fiber sample, fibers much longer than the cut length of 60 mm were found in a tangled position. After recycling, the highest frequency was recorded in the 20–25 mm length class, while before recycling the highest frequency was found in the 40–50 mm length class.



Figure 2. Frequency histograms of hemp fiber length tested (**a**) before recycling and (**b**) after recycling. Figure 3 shows the thickness distribution of hemp fibers tested before and after recycling.



Figure 3. Frequency histograms of hemp fiber thickness tested (a) before recycling and (b) after recycling.

Technical fibers are made up of bundles of elementary fibers (cells) glued together by the middle lamella. During processing, coarse technical fibers are divided into finer fibers. This behavior explains why the average thickness of hemp fibers decreased from $223.5 \pm 101.8 \,\mu\text{m}$ (before recycling) to $174 \pm 77.5 \,\mu\text{m}$ (after recycling), representing a decrease of 22.15%. The recycled fibers had variable thicknesses ranging from 37 μ m to 433 μ m, from fine to coarse fibers. The fiber aspect ratio (length/thickness) of the recycled hemp fiber was 142.5, which is much higher than 10, which is considered the minimum value for a good transmission of stress [81].

Figure 4 shows the images of raw and recycled hemp fibers. In the raw hemp sample, compact fibers (Figure 4a), fibers with split ends (Figure 4b), and fibers with compact ends and split bundles between ends could be found (Figure 4c). The recycled hemp fibers showed the same appearance as raw hemp fibers (Figure 4d–f).

3.2. Mechanical Properties

The tensile strength and the breaking elongation of the manufactured composites are shown in Figure 5.

As shown in Figure 5a, compared to the tensile strength of composite material V1, which is the reference for comparison, the use of recycled fibers to obtain nonwoven fabrics led a reduction in the tensile strength of the composite materials. Thus, a loading of 50% recycled fibers reduced the tensile strength of the composite material by approximately 17.1% (V3), while a loading of 100% recycled fibers decreased the tensile strength of the composite by 22.6% (V2). This reduction in the tensile strength of the composite strength of the composite with

increasing recycled fiber content can be explained by the deterioration in fiber characteristics that occurred in the opening process of the nonwoven waste. The addition of MAPP compatibilizer resulted in an increase in tensile strength of both V2 and V3 composite materials. Thus, in the case of the V2 composite material, the addition of 2.5% MAPP led to an increase in tensile strength of 6.2%, from 20.8 MPa to 22.1 MPa (V2.1), while the incorporation of 5% MAPP caused a higher increase of 19.2%, from 20.8 MPa to 24.8 MPa. This increase in tensile strength of composites treated with MAPP can be explained by the improvement in the interfacial adhesion between the reinforcing hemp fibers and the rPP matrix [82] due to the formation of covalent ester bonds between the anhydride groups of MAPP and the hydroxyl groups of the hemp surface [83].



Figure 4. Images of raw hemp fibers (a-c) and recycled hemp fibers (d-f) at $128 \times$ magnification.



Figure 5. Tensile strength (a) and breaking elongation (b) of composite materials.

Figure 5b shows the breaking elongation of the investigated composite materials. It can be seen that the use of recycled fibers causes a decrease in the elongation at break of the composite materials. Compared to the elongation at break of the V1 composite, the elongation at break decreased by 12.4% for the V3 variant (50% recycled fibers) and by 20.1% for the V2 variant (100% recycled fibers). An explanation for this behavior may be the deterioration of the elasticity of the fibers caused by the mechanical actions to which they were subjected during defibration of the nonwoven waste [84]. The elongation at break of composite materials increased along with the increase in MAPP content. This result is consistent with the findings of Saad [83] and can be explained by the plasticizer role of MAPP [85,86]. Plasticizer separates the rPP chains and reduces the intermolecular forces between them. Thus, the rPP chains can move more easily in relation to one another [87]. Chun et al. reported a decrease in the elongation at break of composites along with an increase in the MAPP content due to the mobility restriction of the PP chains as a result of the improvement in the interfacial adhesion between the matrix and the reinforcing fibers [88]. They also noticed the plasticizing effect of MAPP at concentrations higher than 5%.

The flexural strength of the composite materials is presented in Figure 6a. The reincorporation of recycled fibers into nonwovens leads to a decrease in the flexural strength of the composites, mainly due to the reduction in the length of the hemp reinforcing fibers induced by nonwoven waste reprocessing. Thus, the incorporation of 50% recycled fibers diminished the composite flexural strength by approximately 6.6% (V3), while incorporation of 100% recycled fibers reduced the composite flexural strength by 9% (V2). Adding MAPP to the composites improved the interfacial bonding between the reinforcing hemp fibers and the polymer matrix, and this led to an increase in flexural strength of the composites. However, the increase in flexural strength of MAPP-treated composites was less than 4%.



Figure 6. Flexural strength (a) and flexural modulus (b) of composite materials.

Figure 6b presents the flexural modulus of the analyzed composites. As the content of the recycled fibers into composites increases, the flexural modulus decreases due to the deterioration of fiber characteristics during reprocessing. The flexural modulus of V3 (50% recycled fibers) and V2 (100% recycled fibers) composite variants decreased by 10.3% and 37%, respectively, compared to the flexural modulus of the V1 variant. The addition of MAPP improved the flexural modulus of the composites containing recycled fibers (+14.8% maximum increase for the V2.2 variant) without reaching the value of the flexural modulus of the V1 composite material (4045 MPa).

3.3. Thermal Properties

The thermal behavior of rPP, hemp (H), and the obtained composite materials was studied by thermogravimetry. The analyzed samples were subjected to a dynamic heating cycle (according to Section 2.4). The obtained thermogravimetric curves are shown in Figures 7–9.



Figure 7. Graphs of thermogravimetric curves (**a**) and DTG (**b**) obtained for hemp fibers, rPP, and composite materials.



Figure 8. Graphs of thermogravimetric (**a**) and DTG (**b**) curves obtained for the composite materials from V2 variants.



Figure 9. Graphs of thermogravimetric (**a**) and DTG (**b**) curves obtained for the composite materials fromV3 variants.

 T_{onset} (°C), T_{peak} (°C), and T_{endset} (°C) temperatures were automatically determined from the thermogravimetric diagrams obtained with the device using a Mettler Toledo TGA/SDTA 851.

Table 2 shows the main values extracted from these graphs, such as T_{onset} (°C), T_{peak} (°C), T_{endset} (°C), and mass loss (%).

Sample	Stage	T _{onset} (°C)	T _{peak} (°C)	T _{endset} (°C)	Mass Loss (%)
	Ι	30	52	85.05	7.5
Hemp, 100%	II	303	359	377	54.74
	III	377	446	499	11.31
rPP, 100%	Ι	351.5	423.94	448.96	89.33
	Ι	31	61.79	65.81	5.8
V1	II	321	355	369	16.9
	III	405	465.6	483.37	65.08
	Ι	50.43	70.76	103.75	4.7
V2	Π	325.5	355	369.4	13.72
	III	428.67	464.6	484.6	62.54
V2.1	Ι	70.84	75.6	111.65	4.6
	Π	309.4	355.14	412.62	29.58
	III	466.97	507.33	534.31	58.22
V2.2	Ι	26.07	59.62	65.79	4.5
	II	330.91	353.24	365.52	21.13
	III	407.54	432.4	461.89	58.60
V3	Ι	27.71	69.76	113.80	4.6
	II	326.5	355.83	371.62	13.73
	III	419.4	461.7	479.7	70.68
V3.1.	Ι	31.81	70.10	114.40	4.42
	II	334.94	359.66	371.65	15.48
	III	406.81	455.16	478.09	65.89
V3.2.	Ι	33.34	61.11	103.90	4.40
	Π	327.84	359.49	374.28	17.6
	III	431.29	466.72	485.66	63.29

Table 2. Thermogravimetric parameters of fibers and composite materials.

Figures 7–9 present the TG and DTG thermograms of hemp reinforcing fibers, rPP fibers, and composite materials. The TG thermogram of hemp fiber indicates a mass loss of 7.3% for the temperature range of 41 °C to 149 °C. This mass loss can be explained by the moisture evaporation at the surface of hemp fibers. As temperature further increased between 149 °C and 303 °C, small mass losses were registered (7.06%). In the temperature domain between 303 °C and 376 °C, because of the degradation of cellulose and hemicellulose, the highest mass loss of 57.7% was recorded (maximum degradation rate at 359 °C). Above 376 °C, further mass loss was registered because of lignin degradation [89].

In the DTG curve, the peak at 55 °C was caused by hemp fiber moisture evaporation, and the steep peak at 559 °C was caused by the degradation of cellulose and lignin. The peak at 446 °C is associated with degradation of cellulose, lignin, and PP [90,91].

The TG curve of the recycled polypropylene fiber shows a maximum degradation of the fiber in the temperature range of 351 °C to 448.96 °C, with a mass loss of 98.14%. The DTG curve indicates that the maximum rate of degradation of the fiber took place at a temperature of 423.94 °C.

During the heating process, the transformations of composite materials take place in three stages. In the first stage, the moisture evaporates from the composite material. The highest mass losses were recorded for the V1 variant (5.8% in the temperature range of 31–65.8 °C, with a maximum at 61.7 °C). For the other materials, the mass losses were very close—between 4.7% (for V2 variant) and 4.4% (for V3.2 variant)—and the temperatures did not vary much, ranging between 61.79 °C (for V1) and 59.62 °C (for V2.2).

In the second stage, the temperatures at which degradation was maximum were around 355°C for all the analyzed materials, and the mass loss was between 13.73% for the V3 variant and 29.58% for the V2.1 variant.

In the third stage, the mass loss was high, ranging between 58.22% (for the V2.1 variant, reaching a maximum at 507.33 °C) and 70.68% (for the V3 variant at the maximum temperature of 461.7 °C).

Mass losses were higher for the V3 variant (50% recycled fibers) compared to the V2 variant (100% recycled fibers) (Figures 8 and 9 and Table 2). The addition of the MAPP compatibilizer led to a slight decrease in the thermal stability of the composite material.

The analysis of DTG curves indicates that the V1, V2, and V3 composite materials showed a similar behavior. The first stage in the range of 54–120 °C corresponds to the loss of moisture, the peak at 355 °C corresponds to the degradation of hemp, the peak at 393 °C can be assigned to the degradation of lignin, and the peak at about 465 °C corresponds to rPP degradation. The temperature range indicated for the use of this composite is 20–120 °C, where mass losses are minor.

DSC analysis allowed for the determination of crystallization temperature (T_c), crystallization enthalpy (ΔH_{fc}), melting temperature (T_m), and melting enthalpy (ΔH_{fm}) of the investigated samples. The obtained data are presented graphically in Figures 10–12 and Table 3.

The total crystallinity of the composite material was calculated with Equation (1). Analysis of the data presented in Table 3 indicates that:

- The melting temperatures of the composite materials containing recycled fibers (V2 and V3) are higher than the melting temperature of the V1 composite material and lower than the melting temperature of the rPP matrix;
- With the addition of a compatibilizer, the melting temperatures decreased slightly along with the increase in the amount of MAPP;
- The crystallization temperatures of V2 and V3 composites are lower than the crystallization temperature of V1 composite material and higher than that of the 100% rPP matrix;
- The difference between the crystallization temperature of rPP and the crystallization temperatures of the obtained composite materials is approximately 4.5 °C, varying between 117.54 °C and 122.25 °C;
- With the addition of a compatibilizing agent (MAPP), the crystallization temperatures
 of the obtained composite materials show a slight increase compared to the untreated
 materials; the crystallization temperature also increases along with the increase in
 the concentration of the compatibilizer. This fact confirms that the hemp fibers acted
 as nucleating agents, and consequently, the rPP in the composite materials began to
 crystallize at temperatures higher than 117.54 °C, with the surface of the hemp fibers
 constituting crystallization centers for the polymer matrix [92].

In all analyzed cases, the crystallinity of the composite materials was lower than the crystallinity of the rPP matrix.

3.4. Surface Morphology and Chemical Analysis (SEM and EDX)

Different magnitudes were used to capture images of the composite materials. At $100 \times \text{magnification}$, the images show a multidirectional orientation of the reinforcing hemp fibers in the composites (Figure 13a). Both the way the nonwovens were layered in the mold and the random orientation of the hemp fibers in the nonwovens give the composites fairly similar mechanical properties, regardless of the direction of the applied stress. Furthermore, as indicated by Figure 13a, the hemp fibers are uniformly distributed and well-embedded in the polymeric matrix. Some regions with poor adhesion of the matrix to the fibers could be observed, especially in the case of composites that were not treated with MAPP (Figure 13b). Such local fiber–matrix detachment may affect the structural strength of the composites.

The MAPP-treated composites show a good coverage of the reinforcing fibers and a good adhesion at the fiber–matrix interface (Figure 13c). Some images showed the presence

of microvoids in the matrix structure (Figure 13d). Due to the hydrophilicity of hemp fibers, water molecules inside the microfibrils may gasify at the high temperatures of the forming process and produce voids in the composite material [93]. As shown by the TG thermogram the hemp fiber, a mass loss of 7.3% occurred due to moisture evaporation. The presence of voids can result in the degradation of the mechanical properties of the composites.



Figure 10. DSC (heating process) curves (**a**) and DSC (cooling process) curves (**b**) for hemp fibers, rPP, and composite material variants.



Figure 11. DSC (heating process) curves (**a**) and DSC (cooling process) curves (**b**) for V2 composite material variants.

One of the most effective ways to improve the polymer matrix–reinforcement material interactions is the use of coupling agents. Functionalization with MAPP facilitates the chemical reactions between components such as polypropylene and hemp fibers (used as reinforcements), increasing the adhesion at the interfaces and implicitly improving the mechanical and thermal properties of the composite due to the formation of new bonds between the components. [82,94]. The reactions that take place are shown schematically in Figure 14.



Figure 12. DSC (heating process) curves (**a**) and DSC (cooling process) curves (**b**) for V3 composite material variants.

Sample	First Heating		Cooling		Second Heating			
	Τ _m (°C)	* ΔH _{fm} (J/g)	Τ _c (°C)	ΔH _{fc} (J/g)	Хс (%)	Τ _m (°C)	** ΔH _{fm} (J/g)	Хс (%)
rPP 100%	156.52	49.53	117.54	93.73	45.28	168.87	81.82	39.52
V1	164.22	39.55	122.04	40.06	19.35	163.89	42.05	20.31
V2	164.20	47.81	120.69	57.47	27.76	165.10	51.22	24.74
V.2.1	164.37	34.57	121.07	39.16	18.92	163.37	32.57	15.73
V.2.2	162.89	27.87	121.69	38.04	18.76	162.22	30.07	14.53
V3	165.47	43.05	119.94	51.96	25.10	165.81	48.97	23.65
V.3.1	165.49	38.06	120.79	45.68	22.06	165.49	40.76	19.69
V.3.2	165.28	48.57	122.25	57.27	27.67	164.11	51.96	25.10

Table 3. DSC data for rPP and hemp/rPP composite materials.

* First heating; ** second heating.

The results of the EDX analyses for the examined samples are shown in Figure 15. Five determinations were made at various points on the surface of each analyzed composite material. The average values of the weight percentages (Wt.%) and the atomic percentages (At%) of the elements from each analyzed sample were determined.



Figure 13. SEM images of (a) V1 composite $(100 \times)$, (b) V2 composite $(1000 \times)$, (c) V 3.2 composite $(200 \times)$, and (d) V3 composite $(2000 \times)$.



Figure 14. Scheme of the reaction between the hydroxyl groups on the surface of the hemp fiber, rPP, and MAPP.

The higher values of the O/C ratio for the samples treated with MAPP (Figure 14b,c,e,f), which increase along with the increase in MAPP concentration, confirm the presence of the compatibilizer in the analyzed composite materials.



Figure 15. EDX elemental analysis of composite materials V2 (a), V2.1 (b), V2.2 (c), V3 (d), V3.1 (e), and V3.2 (f).

4. Conclusions

The increasing use of composites in the furniture industry has led to the need to consider the environmental issues generated by waste disposal in the new product develop-

ment process. Waste recycling reduces the environmental impact of waste disposal, lowers the cost of products, conserves resources, and closes the loop in the circular economy.

This research investigated the possibility of recycling hemp/rPP nonwoven waste resulting from the cutting stage in the manufacturing process of 3D composites for upholstered furniture products. Nonwoven scraps were defibrated, and the resulting recycled fibers were reincorporated into new nonwovens from which composites were obtained by thermoforming. The recycling process of nonwoven waste resulted in a 57.09% reduction in hemp fiber length and a 22.15% reduction in hemp fiber thickness. The effect of recycled fiber content (50% and 100%) and MAPP content (2.5% and 5%) on the mechanical and thermal properties of composite materials was studied. This study showed that the mechanical properties of composite materials decreased with increasing recycled fiber content and increased with increasing MAPP content. Thus, the incorporation of 50% and 100% recycled fibers into composites decreased the tensile strength by 17.1–22.6%, the elongation at break by 12.4–20.1%, the flexural strength by 6.6–9%, and flexural modulus by 10.3–37%. The addition of 5% MAPP showed the greatest improvements in mechanical properties for composites containing 100% recycled fibers, as follows: 19.2% increase in tensile strength, 3.8% increase in flexural strength, and 14.8% increase in flexural modulus. Thermogravimetric analysis of the composite materials indicated that the recommended temperature range for composite applications is 20–120 °C, where mass losses are minor. The incorporation of recycled fibers from nonwoven waste into composites leads to an increase in the melting temperature and a decrease in the crystallization temperature. The crystallinity of the composite materials is lower than the crystallinity of the rPP matrix. SEM images prove a good adhesion between the rPP matrix and hemp reinforcement in the case of composite materials treated with MAPP. EDX analyses confirm the presence of the compatibilizing agent in the structure of the treated composite materials.

Composite materials containing up to 50% recycled fibers from nonwoven waste treated with 5% MAPP can be used successfully in furniture applications.

Author Contributions: All authors contributed to the research presented in this work. Their contributions are presented below. Conceptualization, methodology, and investigation: M.I., E.I.M., I.F. and L.S.; formal analysis and data curation: M.I., E.I.M., L.S. and G.L.; writing—original draft preparation: M.I. and E.I.M.; funding: I.F.; project administration: I.F.; writing—review and editing, M.I. and E.I.M. All authors have read and agreed to the published version of the manuscript.

Funding: Publication fees were financed by the Gheorghe Asachi Technical University of Iași, Romania from the Fund for University Scientific Research.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: All data are included in the published article.

Acknowledgments: This work was developed within the framework of COP Project No. 267/22.06.2020, MySMIS 121434, "Creating a center of excellence in the field of composite material at SC TAPARO SA", funded by the European Union via the European Regional Development Fund and the Romanian Government.

Conflicts of Interest: The authors declare no conflict of interest.

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