ECO-FRIENDLY COMPOSITE FABRICATED FROM RECYCLED WASTE PAPER AND RECYCLED POLYPROPYLENE

JEVGENIJS JAUNSLAVIETIS,[*,**] GALIA SHULGA,[*] MARTINS KALNINS,[**] JURIS OZOLINS,[**] BRIGITA NEIBERTE,[*] ANRIJS VEROVKINS,[*] LAIMA VEVERE[*] and TALRITS BETKERS[*]

[*]*Latvian State Institute of Wood Chemistry, 27 Dzerbenes Str., LV-1006, Riga, Latvia*
[**]*Riga Technical University, 3/7 Paula Valdena Str., LV–1048, Riga, Latvia*
✉ *Corresponding author: G. Shulga, galshulga@inbox.lv*

The goal of the work was to recycle waste paper, utilize it as a filler and investigate the properties of the obtained recycled polypropylene-based composite. Crystalline cellulose from the waste paper was regenerated in accordance with the developed method in acid medium under mild conditions. The morphology, crystallinity degree, shape, and sizes of the obtained cellulose particles were characterized. It was found that the extracted crystalline cellulose contained more amorphous regions than the kraft pulp, and was characterized by a more hydrophilic structure. The extracted crystalline cellulose microparticles were used as a biofiller in the recycled polypropylene-based composite in its binary and ternary formulations, incorporating maleic anhydride-grafted polypropylene as an adhesion enhancer. The binary composite was characterized by unsatisfactory mechanical and wetting properties. The presence of the adhesion enhancer in the composite's formulation improved the mechanical properties, increased the hydrophobicity and thermal stability of the composite.

Keywords: waste paper, microcrystalline cellulose, composite, contact angle, surface free energy, thermal stability, X-ray diffraction

INTRODUCTION

The recycling of paper and polymer wastes has significantly improved in recent times, but a large part of them is still burned or sent to landfills. Landfilling and burning of these wastes cause serious environmental problems because during these actions toxic and carcinogenic substances, including carbon dioxide, are emitted into the environment and lead to climate change. Reducing the concentration of atmospheric carbon dioxide is a fundamental challenge for the 21st century. The principles of circular economy impose the need to manage municipal wastes more efficiently by creating eco-friendly materials and marketable end products through innovative technologies. The recycling of waste paper to obtain cellulose has a positive impact on the environment and has economic benefits. Modelling the evolution of the global waste paper trade shows that the global volume of waste paper in the last 25 years reached 1010 million tons.[1] The majority of waste paper is used in papermaking to produce cellulose, with one ton of waste paper saving nearly 17 trees and 7000 gallons of water and decreasing nearly twice the cost of the produced pulp.[2]

Obtaining cellulose from paper wastes can be regarded as one of the promised options for their recycling. The extraction of cellulose from lignocellulosic biomass can be performed not only by pulping, but also by its mechanical disintegration, reactive extrusion, steam explosion, and alkaline and acid hydrolysis.[3-6] The treatments destroy the amorphous structures and leave the crystalline domains in cellulose. Each of the used extraction methods of cellulose from biomass has its own advantages and drawbacks. It should be noted that crystalline cellulose obtained by various extraction methods is characterized by different values of crystallinity, particle sizes, surface area, porosity, moisture content, *etc*. Acid hydrolysis is one of the most popular methods for obtaining crystalline cellulose from lignocellulosic biomass, such as newsprint,[7] filter paper,[8] cotton wastes,[9] straw,[10] hemp stalks[11] *etc.* Acid hydrolysis can be carried out using inorganic acids, such as sulfuric, hydrochloric,

hydrobromic, as well as ionic liquids. The attractiveness of crystalline cellulose for obtaining different functional and high-value-added products is conditioned by its properties, *i.e.* strength, stiffness, water insolubility, non-toxicity, biodegradability and renewability. Due to these properties, crystalline cellulose derived from the waste biomass can be applied in various fields,[12] including the fabrication of wood-plastic composites (WPCs).[13-16]

Polypropylene is one of the most recyclable materials, and in fact, in 2006, the amount of recycled plastics was estimated to be around 4.4 million tons of plastics.[17] Recycling PP has environmental benefits, because it diminishes the plastic waste volume and decreases the carbon dioxide concentration in the atmosphere due to the absence of PP waste burning. The mechanical process of recycling PP involves several stages, namely, PP wastes collection, sorting and cleaning, shredding, separation and compounding. The obtained waste plastic mass is processed using an extruder, which melts the plastic mass at a high temperature, creating recycled PP granules. Due to their large volume and low cost of polymer wastes, their use as a polymer matrix in WPCs is a promising way for rational recycling and reducing their load on the environment.[18,19] WPCs are a young generation of composites with rapidly growing usage within the plastics industry. This type of composite materials belong under eco-friendly composites as the content of the lignocellulosic filler in them can reach 70-80 wt%. The WPC global market is increasing at a very rapid pace and is predicted to exceed 10 billion dollars by 2026. This type of composite finds wide applications for producing decking and decorative materials for building engineering, in the auto industry, for the production of domestic and technical items, *etc*.

The main challenge of WPCs is insufficient adhesion at the lignocellulosic filler/polymer matrix interface that negatively affects the WPC properties.[20,21] Strong interfacial adhesion is needed for the prevention of cohesive fibre-fibre interaction, leading to agglomeration of lignocellulosic particles. The improvement of the adhesion affinity between a polyolefin and a lignocellulosic filler can be achieved by the application of specific functional additives, such as compatibilizers, coupling agents and adhesion enhancers[22,23] in WPCs. However, there is no strict nomenclature designation for these additives yet. The functional additives increase the interface adhesion and decrease the particle agglomeration due to their physicochemical interactions with the components of WPC. The most known maleic anhydride-grafted polyethylene (PE-g-MA), maleic anhydride-grafted ethylene octane copolymers (EC-g-MA) and maleic anhydride-grafted polypropylene (PP-g-MA) are often used in formulations of polyolefin-based composites.[24] The goal of the present work was to recycle waste paper using the developed method for cellulose regeneration and characterize the recycled product, then use it as a biofiller in a recycled polypropylene-based composite and study the properties of the obtained composite samples.

EXPERIMENTAL

Materials

Biofiller

The waste paper represented a de-inked newspaper. For obtaining the biofiller, a new method for its recycling was proposed. The waste paper was cut into small pieces (1×1 cm), which were soaked in 0.05% HCl solution (waste paper/solution = 1/20) for 60 min. Then, the excess water was removed, and the soaked waste paper was dried in 2 steps. First, it was dried for 16 h at 60 °C, then for 2 h at 120 °C. A photo of the dried recycled waste paper (C-WP) before milling in the planetary ball mill is shown in Figure 1.

Figure 1: Photograph of treated deinked waste paper before milling

Milling of the dried waste paper was performed in a planetary ball mill (Retsch, Germany) for 10 min at 300 rpm. Then, the resulting powder was washed until neutral pH and dried again. The obtained recycled waste paper was characterized by elemental and functional analysis, X-ray diffraction method, particle size distribution and scanning electron microscopy. Its surface wettability was also estimated. For comparison, birch kraft pulp (C-P), with microparticles with a size of less than 45 μm, was exploited.

Polymer matrix

Recycled polypropylene (rPP) (0.9 t/m^3, 5.2 g/10 min at 230 °C, and 2.16 kg) was purchased from a recycling plant (Nordic Plast Ltd., Latvia) and used as a polymer matrix. Before blending, it was milled in a knife mill (Retsch, Germany) and sieved through a sieve with a hole size of 500 μm.

Adhesion enhancer

Maleic anhydride grafted polypropylene with the trademark Licocene PP MA 7452, produced by Clariant Company, was used as an adhesion enhancer (AE). Its amount in the composite samples did not exceed 3 wt%.

Composite samples

The composite blend was obtained by mixing the powdered rPP with the milled recycled waste paper, both in the absence and the presence of the adhesion enhancer, for 5 min at room temperature in a Pulverisette 0 vibratory micromill (Frisch, Germany). The cellulosic microparticle content in the blends varied from 10 wt% to 50 wt%. The obtained blends were used for fabricating composite samples using a Haake MiniLab II and MiniJet II (Thermo Fisher Scientific, Germany) at a temperature of 175 °C, a circulation time in the two-screw extruder of 5 min, screw rotational speed of 130 rpm, and a moulding pressure of 60 MPa at a temperature of 120 °C.

Methods
Chemical analysis

Elemental analysis of the recycled waste paper and the birch kraft pulp microparticles was carried out with Elementar Analysensysteme equipment (Elementar Analysensysteme, Germany).

Particle size distribution

The particle size distribution of cellulosic powders was measured using an Analysette 22 NanoTec laser particle sizer (Fritsch, Germany). Before measurements, powder samples were dispersed in water using an ultrasound probe.

X-ray diffraction analysis

XRD analysis of the cellulosic powder samples was carried out using a Panalytical X'pert PRO diffractometer (USA). The samples were scanned over 2θ° range (10° to 90°). The crystallinity index was calculated using Equation (1):[25,26]

$$C_{ri} = [(I_{cr} - I_{am})/I_{cr}] \times 100\% \qquad (1)$$

where C_{ri} – crystallinity index, %; I_{cr} – intensity value for crystalline cellulose (2θ = 22.5°), I_{am} – intensity value for amorphous cellulose (2θ = 18°).

Cellulosic particle sizes

The size distribution of the cellulosic powder particles was determined by an Analysette 22 NanoTec laser granulometer (Frisch, Germany). Before taking a measurement, powder samples were dispersed in water by an ultrasound probe.

Wetting properties

To study the wetting properties of the cellulosic microparticles and the composite samples based on rPP, Washburn and Wilhelmy's methods were used, respectively. To study contact angles, composite samples with dimensions of 60 × 10 × 1 mm were prepared by injection moulding. These samples were conditioned for 24 h at 60 °C before measuring contact angles with Kruss K100M (Kruss, Germany). Capillary constants of the cellulosic particles, conditioned at the same parameters, were found using n-heptane, and then contact angles were measured in water, DMSO, and diiodomethane, The tests were repeated by three times and the obtained results represented average arithmetic values of the measured contact angles.

Work of adhesion was calculated using next equation:

$$WA = \sigma (1 + \cos\Theta) \qquad (2)$$

where WA – work of adhesion, mN/m, σ –surface tension, mN/m, Θ – contact angle,°.

The total surface free energy (SFE) and its dispersive (Lifshitz-van der Waals interactions) and polar (Lewis acid-base interactions) components were calculated using the Owens-Wendt-Rabel-Kaelble method.[27] SFE was calculated using special software installed in the Kruss K100M tensiometer.

Mechanical tests

Mechanical properties of the composite samples were determined with a "Zwick" universal machine (Zwick/Roell, Germany), with a load capacity of 0.5 kN at a rate of 50 mm/min and 2 mm/min for tensile and bending tests, respectively, according to ASTM D638 (2007) and ISO 178 (2010), with the help of the TestXpert I software program (Zwick/Roell, Germany). "Dogbone" samples for tensile tests had the following sizes: overall length of 63.0 mm, overall width of 9.4 mm, length of a narrow section of 9.4 mm, width of a narrow section of 3.15 and thickness of 3.15 mm. For the three-point bending test, samples with a length of 80.0 mm, width of 10.0 mm and thickness of 4.0 mm were used. Five replicates were

made for each mechanical test, and the standard deviations for the values were determined.

Thermogravimetric analysis

TGA and DTG analyses were performed using a TA Instruments Discovery TGA 5500 thermogravimetric analyser. Weight loss was determined in Pt sample pans under an N_2 purge at 50 mL min^{-1} by isothermally treating the sample at 30 °C, followed by heating to 700 °C at a rate of 10 °C min^{-1}. Three replicates were done for each sample.

Scanning electron microscopy (SEM)

The microstructure of the samples was studied using a Tescan Mira/LMU device. Samples were coated with a 15 nm thick gold layer before analysis using Emitech K550X. SEM images were obtained at ×100 and ×5000 magnifications.

RESULTS AND DISCUSSION
Recycled waste paper
Physico-chemical characteristics

The recycled waste paper was obtained using the thermochemical treatment of a deinked newspaper under mild conditions. According to the elemental analysis, C-WP and C-P had the following elemental composition: carbon – 44.86% and 45.53%, hydrogen – 4.47% and 4.49%, oxygen – 50.52% and 49.88%, nitrogen – 0.15% and 0.10%, respectively.

The C-WP particle size distribution reveals (Table 1) that more than 99% of the cellulosic particles were smaller than 200 μm, wherein 19% were between 100 and 200 μm, and 80% of the particles were smaller than 100 μm.

The morphology of the resulting C-WP cellulose microparticles was estimated by the SEM method at different magnifications. It can be seen from Figure 2 that the obtained C-WP microparticles have, predominantly, a needle-like form (×100 magnification). A single cellulosic microparticle is clearly visible at ×5000 magnification. It is characterised by anisotropy with a low diameter-to-length ratio. The diameter of the microparticle, being evaluated from the given SEM image, does not exceed 15 μm.

The X-ray diffraction method is one of the most powerful techniques that give valuable information about the crystal structure of cellulosic materials. According to Figure 3, the obtained XRD patterns for the C-WP and C-P microparticles have different intensities of the peaks in the amorphous region ($2\theta \approx 18.5°$) and crystalline region ($2\theta \approx 22.5°$). Based on the obtained patterns, the crystalline index values, calculated for C-WP and C-P, were 61% and 81%, respectively. These results indicated that the proposed thermochemical treatment could effectively extract cellulose and destroy the amorphous structures in it. At the same time, the content of amorphous regions in C-WP was significantly larger than in the pulp.

Table 1
Size distribution of recycled waste paper microparticles

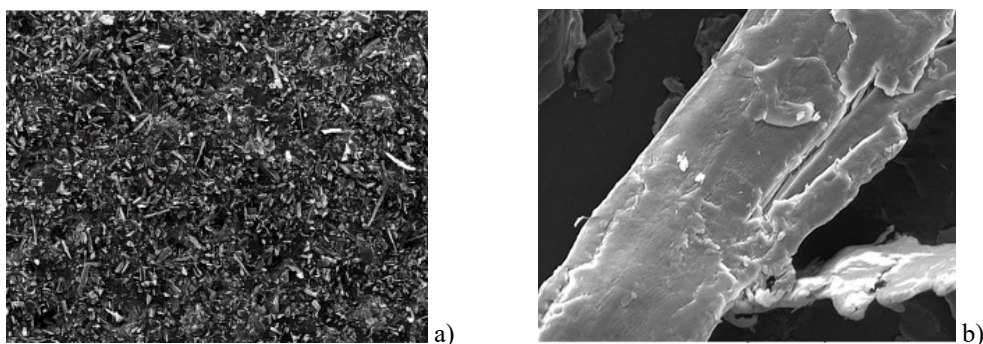| Size, μm	Content, %
> 200	< 1.0
100-200	19.0
< 100	> 80.0

Figure 2: SEM images of C-WP microparticles at various magnification (X100 (a) and X5000 (b), respectively)
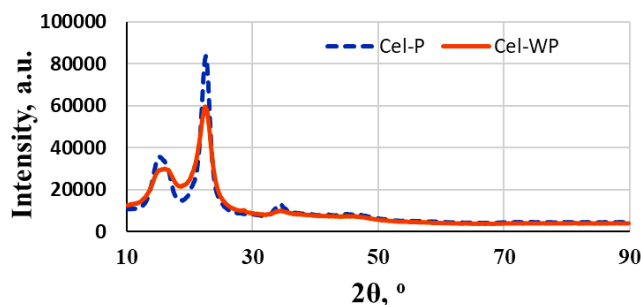
Figure 3: XRD diffraction patterns of C-WP microparticles and the kraft pulp

Table 2
Surface characteristics of recycled waste paper microparticles and kraft pulp

Sample	Contact angles, º			Surface free energy, mN/m
	H_2O	DMSO	DIM	
C-WP	85.6	76.2	63.1	25,3
C-P	85.3	75.5	85.7	12,8

Surface properties

The surface properties of cellulose and cellulose-based materials play an important role in practical application. The influence of the structural properties, particularly, the degree of crystallinity of cellulosic particles on their wetting properties is known. With this aim, the contact angles of the water droplet on the milled recycled paper and the pulp were compared. Table 2 shows the values of the contact angles of the C-WP and C-P microparticles for various liquids, measured by the Washburn method. The contact angles formed between the water droplet and both types of cellulosic microparticles have very similar values. There is not a big difference between the contact angles of C-WP and CP with dimethyl sulfoxide (DMSO). However, there is a noticeable difference in the contact angle values of the compared cellulosic microparticles with diiodomethane (DIM), namely, 63.1° and 85.7° for C-WP and C-P, respectively. A smaller contact angle of C-WP with DIM, in comparison with C-P, is evidently associated with the enhanced content of amorphous regions in its structure, therefore, higher wettability and lower hydrophobicity of the structure.

Surface free energy (SFE) is another important parameter for the characterization of cellulosic materials. SFE of the C-WP and C-P microparticles was calculated from their corresponding contact angles with water, DMSO and DIM. The calculated SFE values are given in Table 2. It can be seen that the total SFE for the C-WP microparticles exceeds almost twice the value for the pulp microparticles, which confirms a more hydrophilic structure of the recycled waste paper, caused by the formation of new hydroxyl and carboxyl groups as a result of its thermochemical treatment.

Composites
Mechanical properties

The blends of rPP and the C-WP microparticles for obtaining composite samples in the absence and presence of the adhesion enhancer were processed in extrusion and injection moulding modes. The content of the initial components in the obtained composite samples is given in Table 3.

Mechanical properties are the most studied material properties due to their importance and ease of testing. It is known that these properties of polymer composites are criteria for evaluating the degree of compatibility between a polymer matrix and a lignocellulosic filler.[24] The mechanical properties of rPP and the rPP-based composite samples filled with C-WP microparticles, with a filling from 10 wt% to 50 wt%, are given in Figures 4-5. The obtained stress-strain dependencies show that, with increasing the content of the cellulosic filler, the values of the mechanical properties of the composite samples deteriorate, except for the mechanical modulus. According to Figure 4, the drop of the tensile strength at break becomes more pronounced at a high filling degree of 50 wt%, being lower by 27% relative to the same parameter of the polymer matrix. The revealed results indicate the deterioration of the tensile properties of the composite samples filled with the C-WP

microparticles, compared with the case of rPP, in spite of increasing the values of Young's modulus up to 22-44% (Fig. 4) and decreasing the tensile deformation values up to 29-300%, for a filler loading in the range of 10-50 wt%. It is known that the increase in mechanical modulus of composite materials with the simultaneous decrease of their deformation is caused by mechanical restrictions due to the presence of filler particles limiting the mobility and deformability of a polymer matrix. The dependences of bending strength and bending modulus on the content of the cellulosic microparticles (Fig. 5) are similar in the case of the tensile properties. With increasing the filler content in the range of 10-50 wt%, the bending strength values at break decreased by 3-29%, but bending modulus values rise by 18-240% in comparison with that of the rPP sample. A decrease in the tensile and bending strength values of the samples filled with the C-WP microparticles is the result of insufficient compatibility between the recycled waste paper microparticles and the polymer matrix[28,29] and

may be caused by the formation of cellulosic microparticle agglomerates due to prevalence of cohesive interactions over adhesive ones.

For improving interface adhesion between the hydrophilic cellulosic filler and the hydrophobic rPP matrix, maleic anhydride grafted polypropylene was introduced as an adhesion enhancer (AE) in the amount of 3 wt% in the composition of the composite samples. Its addition positively affects the mechanical properties of the composite samples. According to Figure 4, with increasing the content of the filler from 10 wt% to 50 wt%, the values of tensile strength and modulus of elasticity for the samples containing AE increase from 21.4 MPa to 30.4 MPa and 966 MPa to 1227 MPa, respectively, which corresponds to an increase in the tensile strength and the modulus of elasticity by 12–58% and 26–60%, respectively, compared with the case of recycled PP. The deformation ability drops by almost 5.5 times for the composite sample with AE at 50% filler loading, relative to the elongation of the polymer matrix.

Table 3
Content of rPP, recycled cellulosic microparticles and adhesion enhancer in composite samples

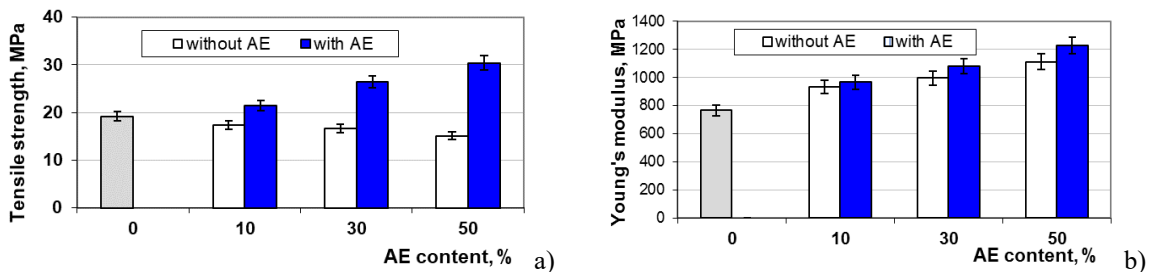| Composition	rPP, %	C-WP, %	AE, %
rPP	100	0	0
rPP/10 C-WP	90	10	0
rPP/30 C-WP	70	30	0
rPP/50 C-WP	50	50	0
rPP/10 C-WP/AE	87	10	3
rPP/30 C-WP/ AE	67	30	3
rPP/50 C-WP/AE	47	50	3

Figure 4: Tensile strength at break (a) and Young's modulus (b) of the composite samples without and with AE adhesion enhancer
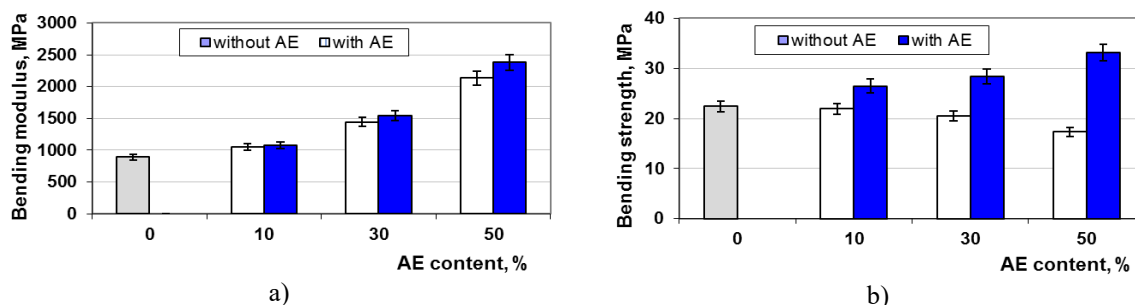
Figure 5: Bending strength at break (a) and bending modulus (b) of the composite samples without and with adhesion enhancer

The values of bending strength and bending modulus for the samples with AE (Fig. 5) boost with increasing the filling degree and are higher by 48% and 2.7 times at the highest filling degree, respectively, but the deflection ability of the samples containing AE diminished more than 4 times at this filling in comparison with the case of rPP. The high values of both elasticity and bending modulus indicate the increased stiffness of the composite samples filled with the C-WP microparticles. Thus, the obtained results show the effectiveness of the applied adhesion enhancer in improving the interfacial adhesion between the recycled waste paper microparticles and the polymer matrix. It is suggested that the boost in the interface adhesion in the developed composite samples, in this case, relates to the formation of physicochemical bonds between the cellulosic microparticles and rPP due to the interaction of AE with both components. This interfacial adhesion enhancement contributes to a more efficient transfer of the load from the polymer matrix to the crystalline cellulosic microparticles.

Thermogravimetric analysis

Thermogravimetric analysis was used to evaluate the effects of the adhesion enhancer on the thermal stability of the composite. The obtained results are presented in Figure 6 and Table 4. The TGA curve of the C-WP particles in Figure 6 does not show any inflection points, and their DTG curve is characterized by only one peak at 351 °C, corresponding to the maximal rate of the thermal degradation of cellulose. It is known[30] that the thermal decomposition of cellulose is accompanied by the cleavage of glycosidic linkages, dehydration of pyranoses and the formation of low-molecular and volatile products at higher temperatures. The weight loss of the cellulosic microparticles is 46.9% at the maximal

rate of thermal degradation, but at the end of the test – 9.3%. There is one big peak at 470 °C on the DTG curve, attributed to the maximal rate of the degradation of the polymer matrix, with a weight loss of 48.1% and 2.4% at the end of the test. The TGA and DTG curves of the polymer matrix have non-pronounced inflection points and peaks in the range of 265 °C-325 °C that may be due to the presence of the low-molecular fractions in rPP. The thermogravimetric curves of the composite sample filled with 30% C-WP microparticles, in the presence of the adhesion enhancer, are characterized by two inflection points and corresponding peaks. The maximal rate of the degradation of the composite sample is achieved at 472 °C, with a weight loss of 47%. The inflection point on the TGA curve, corresponding to a peak at 265 °C, with a weight loss of less than 8%, is absent on the thermal curves of the cellulosic microparticles, but they are non-pronouncedly present on the curves of the polymer matrix. It may be suggested that the revealed peak may be conditioned by the low-molecular products as a result of the interaction between low-molecular fractions of rPP with the C-WP particles during the processing of the composite blend in the presence of AE. Despite the fact that the content of the C-WP microparticles in the composite is only 30%, its maximal rate of destruction is even slightly higher than for rPP, but the weight loss is 4 times lower than for rPP at 500 °C. In such a way, the obtained result can point out that the adhesion enhancer, by simultaneously forming a physicochemical network with both the recycled polymer matrix by H-bonding and the cellulosic microparticles due to the reaction of esterification between its anhydride groups and hydroxyl groups of the C-WR microparticles, increases the thermal stability of the composite.
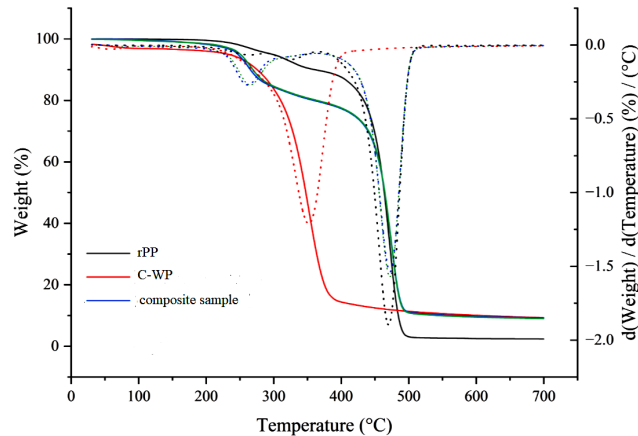
Figure 6: TGA and DTG curves of rPP, recycled waste paper and 30% filled composite in the presence of adhesion enhancer

Table 4
Thermogravimetric analysis of rPP, recycled waste paper and composite sample filled with 30% recycled waste paper

Sample	T_{onset}, °C	T max. rate of weight loss, °C	T max. rate of decomposition, °C	Weight loss, wt%	
				T max. rate of decomp.	Endpoint 500 °C
rPP	265	441	470	48	2.4
C-WP	250	313	351	47	9.4
Composite	265	437	472	47	9.3

Wetting properties

It is known that a major drawback of composite materials containing a cellulosic filler is their pronounced water sorption due to the hydrophilic nature of the cellulosic surface, as well as its roughness and the presence of a porous system. In recent decades, studies devoted to surface energetic characteristics have attracted attention due to the enhanced interest in bio-composite materials. The surface energetic characteristics, such as surface free energy and its components – dispersive and polar parts – could be used to understand and predict the wettability of composites containing a cellulosic filler, because the wettability is one of the most important characteristics for determining their end-use applications and exploitation conditions. The wettability of composite materials depends to a great extent on the interface adhesion quality between the polymer matrix and the cellulosic filler. It was found that the larger the interfacial adhesion, the smaller the wettability of the obtained composite.[28,29,31]

Table 5 shows the values of contact angles and the energy characteristics of the composite samples with different content of C-WP microparticles in the presence of AE. The rPP sample obtained by extrusion and injection moulding is characterized by the highest value of contact angle due to its hydrophobic surface. The introduction of only 10% of the cellulosic filler into the polymer matrix leads to a decrease in the wetting angle of the sample by 4°, which is obviously due to the uneven distribution of the hydrophilic C-WP microparticles in the volume of the polymer matrix. With increasing the C-WP content, the contact angles of the samples enhance and reach values of more than 100°, which is caused by increasing the hydrophobicity of the composite samples in the presence of AE, and, as a sequence, the deterioration in their wettability with water. The growth of the contact angle values of the samples reduces the work of adhesion from 62.3 mN/m to 58.8 mN/m when increasing the filling degree from 10 to 30 wt% (Table 5). According to Table 5, in which the values of SFE and its parts – dispersed and polar – for the composite samples in the presence of AE are reflected, the polymer matrix has the highest value of surface free energy, but the composite sample with a 10% C-WP particle content is characterized by the lowest value of SFE. Comparing the values of the dispersed and polar components of SFE, it can be concluded that the polar part of the polymer matrix is almost 10 times smaller than that of the sample with 10%

filler content. The samples with 30% C-WP microparticle loading have the highest value of dispersive energy and the lowest value of polar energy. The dependence of the SFE polar part of the samples on the content of the C-WP microparticles, in the absence and the presence of AE, is shown in Figure 7. The presence of the adhesion enhancer does not have an essential effect on the polar energy of the samples at a low filling degree. However, by increasing the filler content to 30-50 wt%, the polar energy values of the samples, containing AE, decrease by 2-3 times, compared with the values of the polar part for the samples with the same loading without

enhancer. In the given case, the SFE polar part, characterizing the ability of the obtained composite sample to be wetted, changes more pronouncedly that the values of the SFE dispersive part. It should be noted that the sample with 50% C-WP content has a slightly higher polar energy value than the sample containing 30% filler loading, in the presence of the adhesion enhancer. It can be assumed that this may be due to the presence of partial agglomeration between the C-WP microparticles in the presence of an insufficient amount of AE at the highest filling degree.

Table 5

Contact angle, adhesion work and dispersive part of surface free energy of composite samples with different filling in the presence of adhesion enhancer

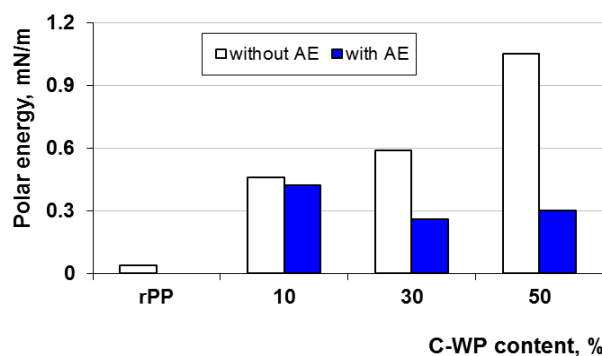| C-WP content, %	Contact angle, °	Adhesion work, mN/m	SFE, mN/m	SFE dispersive part, mN/m	SFE polar part, mN/m
0	102.1	57.3	29.01	28.96	0.05
10	98.1	62.3	27.52	27.08	0.44
30	100.9	58.8	28.49	28.24	0.25
50	99.3	60.3	28.09	27.80	0.29

Figure 7: Polar energy of composite samples without and with adhesion enhancer

Morphological study

The scanning electron microscopy images of cross-sections, obtained as a result of the rupture after the tensile test, for the composite samples with 50% C-WP microparticle content, are shown in Figure 8. The low compatibility between the cellulosic microparticles and the polymer matrix in the sample, in the absence of AE, is reflected by the pronounced heterogenic fracture surface, which is characterised by the formation of cellulosic microparticles pull-out, debonding and the presence of internal voids. These defects are

the main reason for the reduced mechanical properties of the samples in the absence of the adhesion enhancer. On the other hand, the surface of the composite sample containing 3% AE is more homogenous. The presence of structural defects on the cross-section SEM image of the sample is minimal. This confirms the improved compatibility between the crystalline cellulosic microparticles and the polymer matrix in the presence of AE, which is reflected by the enhanced mechanical properties of the composite.
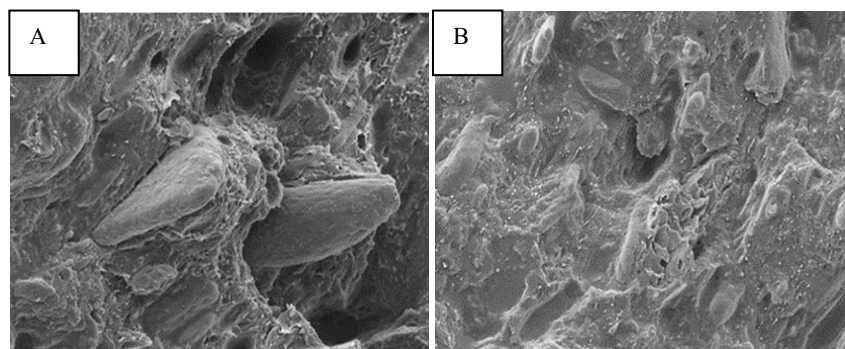
Figure 8: SEM images of the cross-section of 50% filled composite sample (X5000 magnification) in the absence (A) and presence of adhesion enhancer (B)

CONCLUSION

A new technological regime of acid hydrolysis with hydrochloric acid for the extraction of crystalline cellulose from waste paper was proposed. The extracted cellulose was more amorphous than the kraft pulp and had microparticles with sizes below 100 μm. The total surface free energy of the extracted cellulose calculated from the values of the contact angles exceeded almost twice the same value for the pulp microparticles, confirming the enhanced content of amorphous regions in the extracted cellulose, compared with the pulp.

The recycled polypropylene-based composite filled with the extracted cellulose microparticles was fabricated in the absence and presence of maleic anhydride-grafted polypropylene as an adhesion enhancer. An increase in the extracted cellulose content caused a decrease in the mechanical properties and an increase in the wettability of the composite. The addition of the adhesion enhancer to the composite's formulation facilitated an improvement of the mechanical properties and increased the thermal stability. This indicated that the adhesion interaction between the polymer matrix and the extracted cellulosic microparticles prevailed over the cohesive particle-particle interaction. By increasing the cellulose loading to higher values, the polar energy of the composite in the presence of the adhesion enhancer decreased by 2–3 times, compared with the binary composite. Scanning electron microscopy testified that the adhesion enhancer improved interface interactions between the recycled polymer and the biofiller in the composite.

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