MECHANICAL AND MORPHOLOGICAL PROPERTIES OF CELLULOSE-REINFORCED THERMOPLASTIC COMPOSITES: EFFECT OF RECYCLING

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Abstract

In this study, the effect of low microcrystalline cellulose (MCC) content on the mechanical properties of polypropylene (PP) composites when recycled was investigated. The PP/MCC composites were recycled six consecutive times using an injection moulding (IM) machine. Then, the mechanical properties of the composites were examined using tensile and impact testing, Fourier transform infrared (FTIR) spectroscopy, and scanning electron microscopy (SEM). All the mechanical properties of PP/MCC composites, especially the tensile strength (TS), were improved irrespective of recycling rounds. The modulus and impact resistance of the composites were differed slightly after two and four rounds of recycling. The low fibre content and micron-sized fibres did not significantly change the fibre aspect ratio after multiple rounds of recycling. The fracture surface analysis using SEM also reveals that after multiple rounds of recycling, the composites have better cellulose fibre dispersion and good interfacial fibre-matrix adhesion. The results show that the PP composites with low MCC content have good mechanical behaviour. Therefore, recycling PP composites can reduce plastic waste and preserve the environment as well as generate new economic opportunities.

Keywords: Cellulose, Coupling agent, Polypropylene, Recycling, Surface adhesion.

1. Introduction

Plastic waste is a major global contributor to total solid waste in many big cities [1]. The mismanagement of plastic waste as well as the effects of urbanisation and rapid increase in population have caused serious environmental issues because most plastics do not degrade naturally. Recycling is one of the solutions to the environmental problems caused by plastic waste. It also provides an opportunity to create new products that promote a circular economy for plastics. There are two common plastic recycling methods: mechanical and chemical. The mechanical method involves several steps include shredding, re-melting or compounding, and remoulding for product shaping [2] while the chemical method breaks the polymers down into their basic chemicals to produce commodity materials that can be used as feedstock in the polymer industry [3].

The growing awareness of environmental sustainability has driven extensive research in the field of lignocellulosic fibre-reinforced polymer composites, also known as eco-composites or green-composites. Several studies have examined the addition of various types of long, short, micron-sized, and nano-sized lignocellulosic fibres in thermoplastics [4, 5] and thermosets [6, 7]. Flax, kenaf, hemp, jute, empty fruit bunch, cellulose, and wood flour are among the favoured natural fibres. Car interior body components, packaging, construction parts, furniture, musical instruments, and mobile phone casings are some examples of successful eco-composite products available in the marketplace [8]. The lifespans of these products vary according to their applications. For instance, products such as packaging have very short lifespans while construction parts have longer lifespans. Therefore, it is important to recycle polymer composites to reduce the total volume of solid waste because the matrix is usually made of nonbiodegradable plastic, such as PP.

Although the mechanical method of plastic recycling is simple and straightforward, it can degrade the properties of the PP [9]. It has become a tremendous challenge to maintain the properties of recycled PP to ensure that it is still an attractive alternative to virgin polymers. Several studies have examined the effect of multiple rounds of recycling have on the properties of PP-based lignocellulosic composites. Beg and Pickering [10] found that the tensile strength (TS) and Young's modulus (YM) of PP composites filled with 40 wt.% of radiata pine fibre decreased by 25% and 16%, respectively, after the eighth round of recycling. Volt et al. [11] observed similar results after PP/wood fibre composites were recycled nine consecutive times. Both studies observed that these slight decreases were correlated with a decrease in fibre length. Jubinville et al. [12] found that chain scission caused a reduction in the molecular weight (Mw) of PP/wood flour composites that were subjected to thermal-mechanical recycling for six consecutive times. However, it only minimally affected the TS and YM. The repeated recycling also affects the properties of composites from different types of thermoplastic polymer like high density polyethylene (HDPE)/oak wood flour [13] and biomass polymer like polylactic acid (PLA)/wood fibre [14].

All the previous PP composites studies mentioned used high natural fibre content (40 to 70 wt.%). This suggests that the properties of recycled PP-based natural fillers have not been thoroughly explored. Therefore, determining the mechanical properties of PP composites with varying lignocellulosic contents after recycling will provide the industry with useful data in dealing with this material.

The properties of these PP composites can also be tailored or estimated according to the filler content and recycling rounds. Therefore, this study aims to evaluate the effect of recycling on PP composites with low microcrystalline cellulose (MCC) content (5 and 20 wt.%). The mechanical properties were compared and characterised using tensile and impact testing, scanning electron microscope (SEM), and Fourier transform infrared (FTIR) spectroscopy after the second, fourth, and sixth rounds of recycling. The presence of maleic anhydride grafted polypropylene (MAPP) coupling agent in the composites was also examined.

2. Methods

2.1. Materials

Three raw materials were used in this study: (1) recycled polypropylene (rPP) resin, (2) MCC fibre, and (3) MAPP coupling agent. The rPP with a flow index of 1.34 g/10 min and a density of 1.07 g/cm^3 was purchased from Top Flow Industry Sdn Bhd. Both the MCC and MAPP were supplied by Sigma-Aldrich®. The MCC has an average diameter of 20 μm and 3 wt.% MAPP was used.

2.2. Preparation of recycling composites

The first batch of recycled PP/MCC composites were fabricated approximately five years ago. The samples were either dumbbell-shaped type IV or rectangular in accordance with the ASTM D638 and D790 standards, respectively. All these samples were originally prepared for a study by Zulkifli et al. [15]. A twin-screw extruder and an injection moulding (IM) machine were used to compound and fabricate the samples, respectively. The PP/MCC composite samples were prepared by crushing the first batch of samples (5 and 20 wt.% MCC fibres) into small pellets. Then, both samples were processed in the BATTENFIELD® HM 600/850 IM machine. The temperature from 180 to 190 °C was used, while the nozzle temperature was maintained at 200 °C. These steps were repeated multiple times to simulate the recycling process. The mechanical, FTIR, and morphological characteristics of the samples were examined after the second, fourth, and sixth rounds of recycling. The results of the first batch of recycled PP/MCC composite and neat rPP were also compared. Figure 1 depicts the recycling processes of the PP/MCC composites.
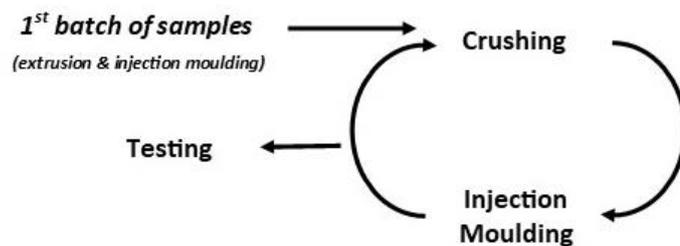

Fig. 1. The multiple round of recycling process of PP/MCC composites.

2.3. Testing and characterisation

Tensile tests were performed at a cross speed of 50 mm/min using the Lloyd Instruments™ Universal Testing Machine according to the ASTM D638 standard.

A minimum of five samples were tested for each composition. The Charpy impact strength test was performed in accordance with the ASTM D256 Method B standard using the Dynisco™ Polymer Test universal impact testing machine. All samples were notched with a mechanical cutter prior to the impact testing.

After the tensile and impact testing were completed, the fracture surface of the samples was coated with a layer of palladium. The JEOL™ JSM 5600 SEM was used to observe the morphology of the fracture surfaces at an accelerating voltage of 7 kV. The structure and functional groups spectra were obtained using the Bruker™ INVENIO® FTIR spectrometer. The spectra recorded were from 4000 to 400 cm^{-1}, with a resolution of 4 cm^{-1}.

3. Results and Discussion

The effect of multiple rounds of recycling on the tensile and impact strength properties and their fracture surfaces, and FTIR were also analysed in this paper.

3.1. Morphological analysis

The fracture surfaces of the samples were examined using SEM to determine the influence of multiple rounds of recycling on samples morphology. Figure 2 shows the tensile fracture surfaces of the PP/5MCC and PP/20MCC samples after multiple rounds of recycling. Plastic yielding was observed in the PP matrix indicating that both samples had fractured due to plastic deformation. Interestingly, the SEM micrographs reveal strong adherence between the MCC fibres and PP matrix, even after the sixth round of recycling (Figs. 2(c), 2(f)).

Here, the MCC fibres remained intact in the PP matrix with plastic yielding features visible around the MCC fibres. Microvoids were also visible in some parts of the fibre-matrix interface due to interfacial debonding. This suggests that multiple rounds of recycling did not deteriorate the MAPP coupling agent. As a result, the load transfer between the matrix phase and the MCC phase during tensile testing was efficient.

The coupling agent also improved the MCC fibre dispersion which reduced the fibre agglomeration. The shear force of the IM machine may also have contributed to better fibre dispersion in the composite during recycling. Figure 2 shows that the sphere-shaped fibre bundles were relatively similar in size of approximately 50 µm; regardless of recycling rounds and fibre content. Small fibre bundles and individual fibres with an average size of 20 µm were still visible (indicated by dotted circles). Meanwhile, the size of the agglomerated MCC fibres in the PP/20MCC sample (Figs. 2(a) to 2(c)) was slightly larger than that of the PP/5MCC sample (Figs. 2(d) to 2(f)) due to the higher MCC fibre content.

Figure 3 shows that there was no apparent plastic deformation or microvoids on the fracture surfaces of the impact testing samples. Prominent differences in the surface morphologies of the PP/5MCC and PP/20MCC samples were also absent. The morphology of the fracture surface was non-uniform and rough in appearance with an irregular patchy structure. Previous studies also described similar fracture surfaces [15, 16]. Each irregular patchy section represents a deformed area and was surrounded by discontinuous edges (depicted by white lines). The surface roughness increased as the MCC fibre content was increased.
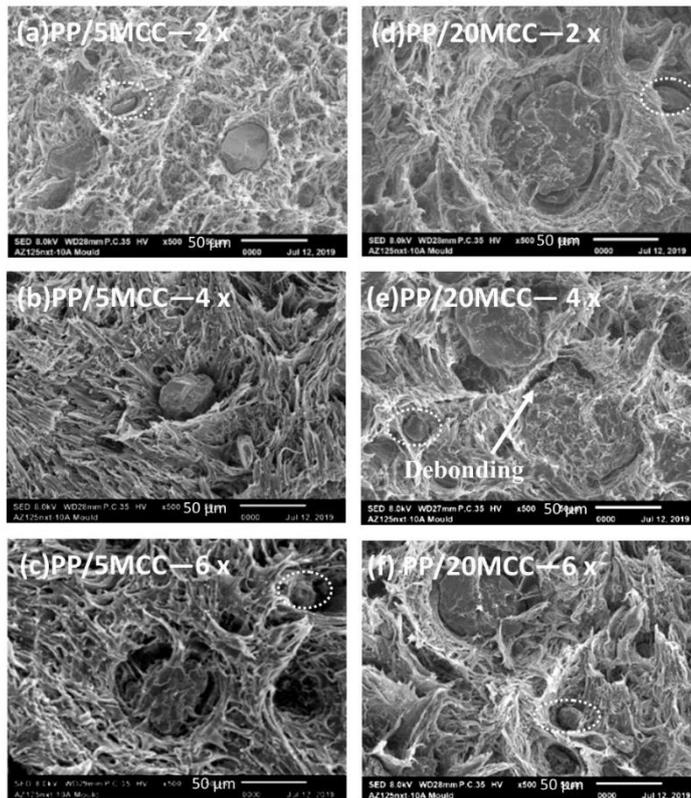
**Fig. 2. SEM micrographs of the tensile fracture surface
of PP/5MCC and PP/20MCC composites after the second (a, d),
fourth (b, e) and sixth (c, f) rounds of recycling.**
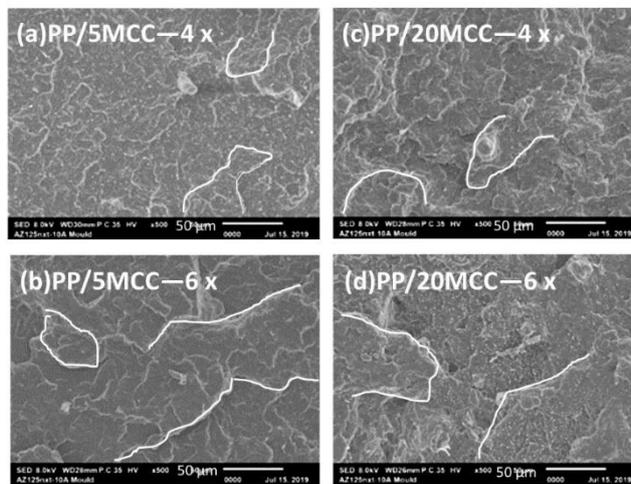
**Fig. 3. SEM micrographs of the impact fracture surface
of the PP/5MCC and PP/20MCC composites after the fourth (a), (c)
and sixth (b), (d) round of recycling.**

3.2. FTIR

Figure 4 shows the FTIR spectra of PP/MCC composites as a function of recycling rounds. As most peaks of neat PP (excluding the ester linkage) are almost similar to the two-round recycling of PP/MCC composites, it was not included in Fig. 4. The broad peak from 3000 to 3600 cm^{-1} represents the OH stretching region and the hydrogen bonds of the hydroxyl groups of the fibre. The ester linkage of MAPP coupling agent between the MCC fibre and the PP matrix was identified by the presence of a small peak at 1732 cm^{-1}. After the fourth round of recycling, a new peak formed from 1670 to 1550 cm^{-1}. This peak was attributed to the C-O bending of the water absorbed by the MCC fibre. It could also indicate the better dispersion of the MCC fibres after multiple rounds of recycling. The peak from 3000 to 3600 cm^{-1} also became broader in the spectrum, indicating that more hydrogen bonds were formed after multiple rounds of recycling. A few peaks of the PP for CH, CH$_2$, and CH$_3$ were observed from 2800 to 3000 cm^{-1}, 1460 to 1450 cm^{-1}, and 1380 to 1370 cm^{-1}, respectively. The peaks at 1167 and 1018 cm^{-1} were attributed to the isotacticity of the PP matrix [17]. A slight decrease in the intensity of PP characteristic peaks indicates that multiple rounds of thermal-mechanical recycling may degrade the composites. It is likely that this degradation also affects the isotacticity of the PP chain because the intensity of these peaks exhibits a similar declining trend. However, no significant carbonyl peaks were noted from 1713 to 1765 cm^{-1}, which is associated with thermo-oxidative degradation [12]. This suggests that six rounds of recycling only resulted in chain scission.
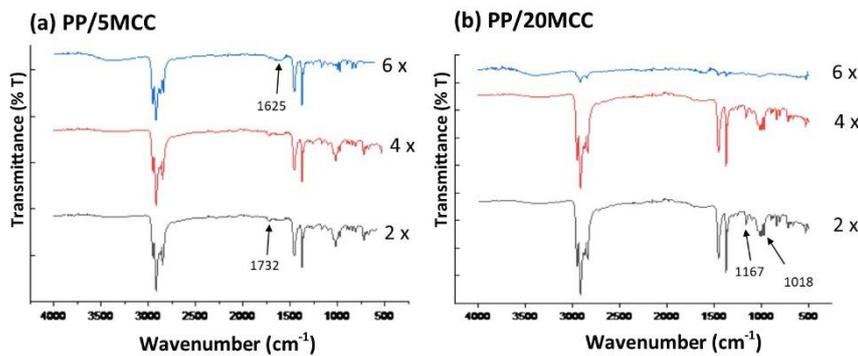

Fig. 4. FTIR spectra of (a) PP/5MCC and (b)PP/20MCC composites at multiple rounds of recycling.

3.3. Mechanical properties

The tensile and impact strength

Figure 5 depicts the YM and TS of PP/MCC composites as a function of MCC content and recycling rounds. A gradual increase in YM was observed after the second round of recycling. However, the YM marginally decreased with subsequent rounds of recycling. The YM of the PP/MCC composites increased as the MCC content was increased. The YM of PP/5MCC and PP/20MCC composites improved by approximately 75% and 112%, respectively, compared to that of the neat rPP. The higher YM of PP/20MCC sample was expected due to the stiffness of the cellulose fibres. The results are also consistent with the rule of mixture.

Although a slight decrease in stiffness occurred after six rounds of recycling, the YM of both composites were still higher than that of the neat rPP, with a decrease of less than 15%.
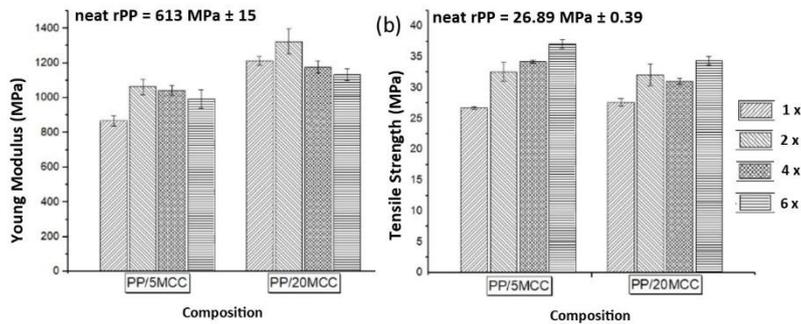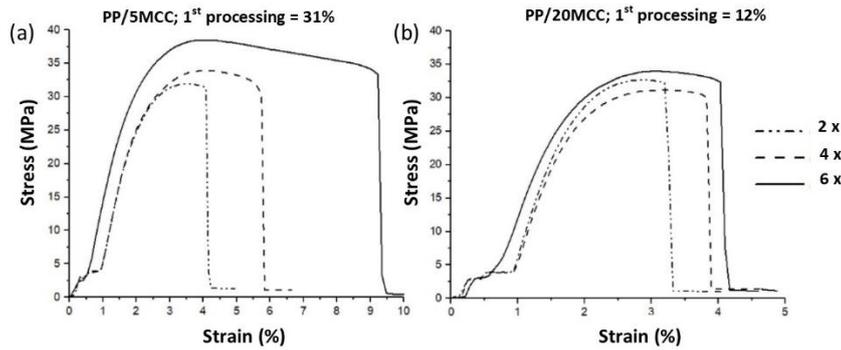

Fig. 5. (a)YM and (b) TS of PP/MCC composites after multiple rounds of recycling.

However, the TS of the PP/MCC composites increased linearly with the MCC fibre fractions at every round of recycling (Fig. 5(b)). The PP/5MCC composite was found to have greater TS than the PP/20MCC composite, especially after the fourth round of recycling. The increase in TS as the recycling rounds number and MCC content increased proved that multiple rounds of recycling did not degrade the function of the MAPP coupling agent in the composite system, as evidenced by the morphological analysis. This is because deterioration of the coupling agent would adversely affect tensile strength [18]. It is noteworthy that the TS results that this present study obtained contradicted the results obtained by Beg and Pickering [10] and Völtz et al. [11] who compared the effects of multiple rounds of recycling on rPP with high reinforcement content and virgin composites. These two extant studies reported that the TS of PP/wood composites was reduced by approximately 21% and 11% after nine and eight rounds of recycling, respectively. The samples used in both studies contained 40 wt.% filler.

Figure 6 illustrates the effect of multiple rounds of recycling on the elongation at break (strain %). The strain % of PP/MCC composite after two, four, and six rounds of recycling was significantly lower than that of the PP/MCC composite after one round of recycling. Interestingly, both samples exhibit a similar trend that the elongation at break increased as the number of recycling rounds was increased. The ductility of PP/5MCC sample was higher than that of the PP/20MCC sample, with a two-fold increase indifference after six rounds of recycling. The low ductility of PP/20MCC sample was due to its higher MCC fibre content, which restricts the deformation of PP matrix. The strain % results are consistent with the findings of previous studies [10, 13].

Figure 7 shows the impact strength of PP/MCC composites as a function of recycling rounds and cellulose loadings. The impact resistance of PP/MCC composites was more than 100% higher than that of the neat rPP. Similar to the TS results, the PP/5MCC composite recorded better impact resistance than the PP/20MCC composite after less than four rounds of recycling. However, the impact resistance of the PP/5MCC composite noticeably decreased after six rounds of recycling.

**Fig. 6. The elongation at break of (a) PP/5MCC and
PP/20MCC composites after multiple round of recycling.**
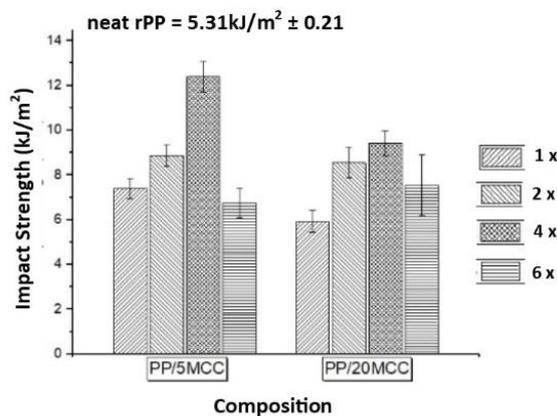
**Fig. 7. The impact strength of PP/MCC
composites after multiple rounds of recycling.**

The mechanical results of this present study varied greatly from that of previous studies that investigated PP composites with high lignocellulosic fibre content [10-12]. These disparities were particularly evident in terms of TS and elongation at break. However, there were some similarities in terms of impact strength and YM. Several possibilities may have contributed to the obtained results. The improved TS obtained by the composites of this present study may be due to the better adhesion of between the PP matrix and MCC fibre interface, as proven by the results of the SEM analysis. Good interfacial adhesion results in better fibre dispersion. Furthermore, coupling agents are known to bridge the gap between the filler and matrix via covalent bonds that facilitate the transfer of stress between these constituents. Sufian et al. [19] and Samat et al. [20] examined PP/alumina and PP/cellulose composite systems, respectively, and found that the tensile properties of composites with coupling agents were higher and vice versa. Apart from good interfacial adhesion, improved tensile properties may also be due to the plastic yielding phenomenon of the PP matrix and interfacial debonding. It has also been suggested that a good fibre-matrix interface

promotes plastic yielding to occur around the fibres. The stresses concentrated around the fibres induce the formation of microvoids via the mechanism of interfacial debonding. As the interfacial adhesion of the composites of this present study was good, debonding occurred only on certain surfaces or at weak points. Furthermore, large-scale plastic deformation and debonding facilitated energy dissipation within the composites during tensile testing.

The improved fibre dispersion within the PP matrix due to multiple rounds of recycling also contributed to the improved TS obtained by the composites in this study. Volt et al. [11] and Soccalingame et al. [21] reported also excellent fibre dispersion as recycling rounds increased. However, increased fibre length has adverse effect on TS because it increased the occurrence of fibre breakage. As seen in Fig. 2, fibre damage occurred less frequently in this present study. Therefore, multiple rounds of recycling did not significantly vary the aspect ratio of the fibres. The size of MCC fibres as well as agglomerated fibres remained consistent. As such, the stability of the aspect ratios of the fibres may also be a factor that affects the mechanical performance of the composites.

The improvements in impact resistance that this present study observed among PP composites with low MCC fibre content that were recycled less than four times are corroborated with the results obtained by Zulkifli et al. [15]. They found that rPP/MCC composites with lower filler content had higher impact resistance due to good filler-matrix adhesion. Due to its ability to yield, the PP matrix was found to govern the toughening mechanism at lower filler contents. Good adhesion further increased the amount of energy required for crack initiation and propagation, which results in toughening during impact. Both conditions are applicable to the findings of this present study for up to four rounds of recycling. However, interfacial adhesion was still present after six rounds of recycling. Thus, the decreased impact resistance could be attributed to the changes in or the degradation of the molecular structure of the PP matrix due to chain scission.

Chain scission is known to affect Mw and viscosity. Viscosity can be determined by analysing the melt flow index (MFI). Generally, an increase in MFI correlates with a decrease in Mw, indicating polymer degradation. Most studies [14, 22] have concluded that multiple rounds of recycling cause chain scission to occur in the amorphous phase of the molecular chain of polymer or PP matrix. A decrease in the length of PP chain is associated with chain scission, which simplifies polymer disentanglement and facilitates polymer mobility [23]. Correlations between an increase in MFI and recycling rounds have been noted in different PP composites systems such as PP/talc [9], PP/natural fibre [10], and PP blends [23]. As seen in Fig. 7, after four rounds of recycling, the PP/20MCC composite had a smaller decrease in impact strength than the PP/5MCC composite. Although chain scission did occur, the higher MCC fibre content, as well as good interfacial adhesion hindered crack propagation in the PP/20MCC composite. On a separate note, elongation at break behaviour at higher recycling rounds are still poorly understood. Better filler dispersion with less agglomeration, good matrix-filler interfacial adhesion, and chain scission may increase elongation percentage. Interestingly, studies on high density polyethylene (HDPE) composites with less than 30 wt.% filler content also reported a similar trend of elongation behaviour after multiple rounds of recycling [13, 24, 25].

Table 1 summarises the results of present study and studies on PP composites reinforced with varying quantities of lignocellulosic fibres. Regardless of processing type, extruder, IM, or a combination, the response of PP composites subjected to an applied load after multiple rounds of recycling depends on the: (i) amount of filler, (ii) fibre aspect ratio, and (iii) coupling agent. Although the mechanical properties of neat rPP decreased proportionally with recycling rounds [16], the PP composites exhibit slightly different behaviour. As seen in Table 1, only insignificant changes in property were observed among the PP composites that were recycled up to five times. Therefore, recycling plastic composites is a sustainable and feasible solution to reduce the total volume of nonbiodegradable waste materials. Furthermore, recycling can be used to produce new products that share similar or different properties than the source PP composites.

Table 1. Mechanical properties PP/lignocellulosic composites.

Composite and filler	CA	RR	YM	TS	Impact strength	Reference
PP/Wood fiber (40-50 wt%) (L=2.36 mm) (D=25 µm)	MAPP (4wt%)	8	40wt%; ↓16% 50wt%; ↑ at 2RR & then ↓	40wt%; ↓25% 50wt%; ↑ at 2RR & then ↓	40wt%; ~48%	[10]
PP/Wood flour (40-70wt%) (Mesh, 60)	-	6	↑ at 3RR & slight different at 6RR	↓ slightly	-	[12]
PP/Wood fiber (40wt%) (L=2-3 mm)	MAPP (% not stated)	9	↑ slightly up to 7RR & ↓ slightly	↓ 21%	-	[11]
PP/Sisal (10-30 wt%) (L=5 mm)	-	4	Not affected	Not affected	-	[26]
PP/MCC (5, 20wt%) (20 µm)	MAPP (3wt%)	6	↑ at 2RR & then ↓ slightly	↑	↑ at 4RR & then ↓	Current work

*↑ - *increase*; ↓- *decrease;* CA - *coupling agent*; RR - recycling rounds number

4. Conclusions

The investigation on the mechanical properties of PP/MCC composites with low filler content after multiple rounds of recycling reveals that:

- Multiple rounds of recycling did not cause a sudden deterioration in the mechanical behaviour of PP/MCC composites with low filler content.
- A few rounds of recycling increased the YM and impact resistance before marginally decreasing while TS continued to increase.
- Multiple rounds of recycling did not severely damage the dimension of the cellulose fibre.
- Multiple rounds of recycling did not degrade the MAPP coupling agent of the composites, as confirmed by FTIR analysis.
- Multiple rounds of recycling and the presence of a MAPP coupling agent improved fibre dispersion as well as interfacial adhesion between the MCC fibre and the PP matrix.

Acknowledgement

Abbreviations	
CA	Coupling Agent
FTIR	Fourier Transform Infrared Spectroscopy
HDPE	High Density Polyethylene
IM	Injection Moulding
MAPP	Maleic Anhydride Grafted Polypropylene
MCC	Microcrystalline Cellulose
MFI	Melt Flow Index
Mw	Molecular Weight
PLA	Polylactic Acid
PP	Polypropylene
RR	Recycling Rounds Number
rPP	Recycled Polypropylene
SEM	Scanning Electron Microscope
TS	Tensile Strength
YM	Young Modulus

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