INTRODUCTION

Polymer composites have been widely used in various applications such as in automotive, aerospace, construction, furniture and packaging. The oil world price crisis and cost effective in the engineering design process has led to the development of eco-friendly materials that is so called green composites [1]. Composite based-natural fiber is also can be categorized as green due to the filler is from the natural source. This composite has received great attention because it is biodegradable, low price, low density and possess good mechanical and thermal properties.

One of the most important polymeric materials for polymer or plastic industry is polypropylene (PP). It offers many advantages such as easy to be processed, low price, high performance and recyclability [2]. Recycling of thermoplastics is important for the sustainability of environment. However, for PP the recycling processes would degrade its properties [3]. The natural fibre is one of the common fillers for polymer composites and one of the components in natural fiber is cellulose. The cellulose may be obtained from plant fiber such as cotton, flax, hemp, jute, sisal, kenaf, coir and palm oil. On the other hand, microcrystalline cellulose (MCC) fiber can be produced from cellulose through acid hydrolysis process [4,5]. During this process the amorphous region would be removed and while the crystalline region remains.

Reinforcement of recycled polypropylene (rPP) with natural fiber likes MCC is an interesting topic. However, very little studies have been carried out to evaluate the potential of MCC as filler for recycled polymers. Hence, the objective of current work was to determine the mechanical and water properties of recycled polypropylene added with microcrystalline cellulose fiber. Since compatibility is crucial issue in polymer based-natural fiber composite, thus maleic anhydride polypropylene (MAPP) would be used as well. The mechanical and water absorption properties of rPP/MCC composites with and without were then compared.

Methodology:

Material and Sample Preparation:

The industrial grade of recycled polypropylene resin was obtained from Sheng Foong Plastic Industries Sdn. Bhd at Tronoh, Perak. It is black in color. Microcrystalline cellulose (MCC) in a form of fine white powder has bulk density of 0.6 g/cm³. Maleic anhydride polypropylene (MAPP) was used as coupling agent with the composition of maleic anhydride, 8-10wt%. It has molecular weight of average Mw~3,900 by GPC ; average Mw~9,100 by GPC and acid number of 47 mg KOH/g. Both microcrystalline cellulose and maleic anhydride polypropylene was supplied by Sigma-Aldrich Co. (M) Sdn. Bhd.

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Composites with various content of MCC (from 0wt% and 40wt%) and a constant amount of MAPP (3wt%) were mixed and compounded with a twin-screw Thermo Haake Rheomexat a temperature of 180°C. Prior to extrusion process, all the raw materials (rPP, MCC and MAPP) were dried at 100°C for 24 hours. The compounding composition is shown in Table 1. The extruded composites were then cooled at room temperature before crushing into pellet. All extruded composites were molded using the Battenfeld HM 600/210 injection molding machine at a pressure of 900 bar and temperatures were kept at from 180 to 190°C and 200°C in the nozzle to produce dumbbell and rectangle shape of samples for testing.

Table 1: The compounding composition of fabricated composites.

<table>
<thead>
<tr>
<th>Material</th>
<th>Sample Without MAPP (wt%)</th>
<th>Sample With MAPP (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MCC</td>
<td>2, 5, 10, 20, 40, 50</td>
<td>2, 5, 10, 20, 40, 50</td>
</tr>
<tr>
<td>MAPP</td>
<td>0</td>
<td>3</td>
</tr>
</tbody>
</table>

21.2 Mechanical Test:

Tensile Test was carried out using a LLYOD Instruments Machine and samples were tested according to ASTM D638. The crosshead speed of testing was fixed at 50mm/min. The average of five samples was assessed each composition. Impact test was performed on the rectangular shape of samples by advanced pendulum impact tester (Dynisco Polymer Test) according to ASTM D256 standard.

1.3 Water test:

The sample of composites with rectangular shape were immersed in distilled water at room temperature with different time interval (12, 24, 48, 72 hours). All the water left on the surface of the sample was then quickly wiped off using tissue paper before weighed. A minimum of 5 samples were tested and the mean was reported. The value of water absorption in percentage after being soaked (M_i) was calculated by using the following equation

\[ M_i (\%) = \frac{W_f - W_i}{W_i} \times 100 \]  

Where \( W_i = \) initial weight  
\( W_f = \) final weight

1.4 Field Emission Scanning Electron Microscopy (FESEM):

The observation of fracture surface morphologies of the composites was employed by using JEOL JSM-6700F Field Emission Scanning Electron Microscopy (FESEM). The specimens were coated with platinum-sputtered in a vacuum chamber with JEOL JFC-1600 Auto Fine coater before observation to avoid charging effect.

Results:

Tensile test:

The variation of tensile strength and modulus of rPP/MCC composites with and without MAPP coupling agent are depicted in Figure 1. In comparing the rPP/MCC composites without MAPP to the neat rPP, the tensile strength decreased along with an increase of MCC content. It is likely that the decrement in the tensile strength is associated with the poor interfacial adhesion between of MCC fiber and rPP matrix.

For rPP/MCC composite with MAPP, it is also noticed that the addition of MAPP did not change much the tensile strength of this composites; moderate increment across all the MCC content. However, the effect of MAPP coupling agent in improving the tensile strength of rPP is important at high amount of MCC fiber (40wt%). As seen in Figure 1 the tensile strength of samples with MAPP increased to about 50% compared to samples without coupling agent.

Impact test:

The impact strength of rPP/MCC composites is shown in Figure 3. The impact strength increased slightly at low content of MCC (2-5wt%) prior to decreased after further amount of MCC was added (10 to 40wt%). Generally, samples which contain MAPP shows higher impact strength than samples without MAPP. Similar to tensile result, an advantage of adding coupling agent was seen to occur at high content of MCC fiber (10, 20 and 40wt%); the impact strength leveled off. But the samples without MAPP continuously deteriorated with increasing MCC content.
Fig. 1: Tensile strength of recycled polypropylene composite with various content of MCC fiber.

Figure 2 shows the effect of MCC fiber on Young modulus of rPP composites with and without MAPP. From this result, it is observed that the influence of MAPP on Young modulus of both rPP composites is not significant; the Young’s modulus increased with increasing MCC content. In fact the difference in Young Modulus for samples with and without MCC fiber is only minor for all composition.

Fig. 2: Tensile modulus of recycled polypropylene composite with various content of MCC fiber.

Fig. 3: Impact strength of recycled polypropylene composites with different content of MCC fiber.

Water absorption test:
The water absorption property for both type rPP composites was also assessed and results are plotted as weight percentage vs time(Figures 4 and 5). Apparently, the incorporation of MCC fiber had caused the water...
absorption property of rPP to increase, more significant in rPP composites without MAPP. On the other hand, for the first 12 hours, at low MCC content (below 20wt%), the water absorption percentages for samples with MAPP are almost similar to neat rPP. And for the next 12 hours, the percentages of water absorption of these samples are still low compared to samples without MAPP for the same filler composition.

![Graph](image1.png)

Fig. 4: The water absorption of rPP composites with MAPP at various amount of MCC.

![Graph](image2.png)

Fig. 5: The water absorption of rPP composites with MAPP at various of MCC content.

Figures 6a and 6b compare the water absorption at low (2wt%) and high MCC content (40wt%), respectively. Regardless to the MCC content, initially both types of samples possess the same water absorption property. However, as the time increases the water absorption changed slightly; higher in samples without MAPP. Indeed, increasing in water absorption property is unavoidable in samples which contain high amount of MCC fiber. But again, as observed in Figure 6b, the presence of MAPP could suppress slightly the absorption of water molecules into the composites.

![Graph](image3.png)

(a)

![Graph](image4.png)

(b)

Fig. 6: The water absorption of rPP composites reinforced with (a) 2wt% and (b) 40wt% of MCC.
Fesem observation:

Figure 7 displays the morphology of MCC fiber. A closer examination of this micrograph revealed that the size MCC fibers are inconsistent. Their diameter range is from 10 to 20μm. Most of the MCC fibers show the rod-like structure appearance and their surfaces are also quite smooth.

**Fig. 7:** FESEM micrograph of microcrystalline cellulose (MCC) fiber.

FESEM was also used to investigate the fracture surfaces of tensile test samples and the micrographs are demonstrated in Figure 8. It is clear that sample with and without MAPP exhibited different fracture surface features. The formation of voids around the MCC fiber is evident on the fracture surface for sample without MAPP. Some MCC fibers also detached from the rPP matrix. All these features signify that the interfacial adhesion between the matrix and filler is poor. In contrast, the MCC fibers are seen to attach well with the rPP matrix, indicating improvement in the interfacial adhesion property.

**Fig. 8:** Tensile fracture surface of rPP/MCC composite (a) without (b) with MAPP coupling agent.

Discussion:

The effect of microcrystalline cellulose (MCC) fiber on the mechanical and water absorption properties of polypropylene (PP) from the recycled source was studied and assessed. The tests were conducted on samples containing 2, 5, 11, 20 and 40 wt% of MCC fiber. Results were then compared with the recycled-PP (rPP) composite system which contains MAPP. Here the MAPP acts as a coupling agent. Without the MAPP (Figure 1), the tensile strength deteriorated after the MCC fiber was added. The tensile strength results in the current study are consistent with previous work on polymer composites system [2,6,5].

However, dissimilar results were obtained in rPP/MCC composites with MAPP. An examination of the FESEM micrographs for samples without coupling agent revealed the debonding of MCC fiber (Figure 8a). It is known that the stress can be transferred effectively from matrix to fiber only if the adhesion between these constituents is strong or vice versa. Thus, it can be said that the deleterious effects in the tensile strength in these composites are mainly associated with the poor interfacial adhesion between the filler and matrix. On the other hand, strong bonding features were observed on the fracture surface of samples added with coupling agent (Figure 8b). And as depicted in Figure 1, the tensile strength of rPP/MCC composite improved in the presence of MAPP coupling agent. Hence, it is clear that the changes in the interfacial adhesion property correspond to
the improvement of tensile strength for samples with MAPP. This is because the existence of MAPP led to the formation of hydrogen bonds with the hydroxyl groups of the MCC fiber [8]. Consequently, the bonding strength between filler and matrix increased and resulted in high tensile strength. According to Qui et al. [2] and Spoljaric et al. [7], the addition of coupling agent reduces the agglomeration or improves the dispersion of cellulose fiber, respectively. Nevertheless, the increment in tensile strength in this study is only moderate. It is presumed that a slight increase in the tensile strength may be attributed to the dissimilar size of MCC fibers as shown in Figure 7.

From Figures 2 and 3, it is noticed that the incorporation of MCC fiber did also produce positive outcome on the mechanical properties of recycled PP (Young’s modulus and impact strength), irrespectively whether the coupling agent is present or not. However, overall the mechanical properties of samples with MAPP are still higher than those samples without MAPP. The increase in Young’s modulus indicates that the stiffness of rPP composites depends on the filler factor or cellulose fiber property. Interestingly, the impact strength showed opposite result to the Young’s modulus; samples with low MCC content (2 and 5wt%) have higher impact strength than those samples with high MCC content. A similar finding was also reported by Samat et al. [9] who studied the effect of coupling agent on the mechanical properties of PP/MCC composites. They found that the impact strength of PP greatly increased with the addition of low amount of MCC fiber (8wt%). Further amount of MCC fibers led to decrement in impact strength of samples without MAPP. A study by Razavi-Nouri M. et al. [6] in PP/chopped rice husk also found a similar impact strength result at high content of filler. Dissimilar to this, at high MCC content, the impact strength for samples with MAPP remains quite constant. From this result, it is likely that the presence of MAPP could prevent severe deterioration in the impact strength particularly in polymer composite added with high amount of very short length fiber (less than 5mm).

Water absorption properties in Figures 4 and 5 showed that the water uptake increases with increasing amount of MCC content. This behavior is contributed from the hydrophilic property and also availability of water residence sites [5] of natural fiber. In addition the porous structure of fibers would also accelerate the penetration of water molecules. FESEM micrograph in previous section exhibited the interfacial adhesion between MCC fiber and rPP is poor. Therefore, the absorbed water molecules may also entrap at the interface between the rPP matrix and MCC fiber. For samples with low amount of MCC fiber (less than 40wt%), the presence of MAPP coupling agent reduced the percentage of water uptake up to 24 hours. This improvement may be associated with the good bonding between the fiber and matrix that reduced the absorption of water molecules into the MCC fibers. On contrary, at higher MCC fiber content, the function of MAPP on reducing water absorption diminished.

Conclusion:

The study shows that the addition microcrystalline cellulose alone only improves the tensile modulus. But the presence of coupling agent in the composites leads to better results in all studied mechanical properties (tensile and impact strength) and also water uptake property. These enhancement is associated with the better interfacial adhesion between the microcrystalline cellulose fiber and recycled polypropylene. However, higher content of microcrystalline cellulose diminish the beneficial effect of coupling agent mainly in the water uptake property.

REFERENCE

