Experimental Investigating Effect of Reprocessing on Properties of Composites based on Recycled Polypropylene

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Abstract—This paper is aimed to provide knowledge and understanding to promote the use of recycled materials via experimental study. In this paper, for the first time, effect of reprocessing on properties of recycled PP/talc composites has been investigated via series of experimental tests, while both virgin and recycled PP were used as comparatives. The materials were reprocessed in two different routes, multiple extrusion cycles and multiple injection moulding cycles. Some materials were taken out for testing during each cycle. The tests include mechanical, rheological and thermal. The results were plotted and discussed, and for the first time, results from the two different reprocessing routes were compared. Some phenomena were observed and fitted in others’ studies and prediction. Also, the complexity of composites made from recycled materials under reprocessing was detected, and in need of further research.

Keywords—reprocessing; recycled; polypropylene; talc-filled; extrusion; injection moulding; chain-scission

I. INTRODUCTION

Due to various reasons, such as pressure from legislations [1~3], saving natural resource (mainly crude oil which plastics are made from), reducing cost, landfill and emission, plastic recycling has attracted a broad interest since 1950s. Even after decades of practices and researches, it still poses a difficult challenge both for industry and for academia. Comparing with landfill, incineration for energy recovery and chemical recycling, mechanical recycling has been proved to be the most viable way to deal with the challenge, for it reduces the consumption of natural resources as well as a reduced landfill and material cost [4]. It was estimated that using recycled plastics could reduce greenhouse gas emissions by about 80% [5]. But when compared to virgin plastic, recycled plastic tend to show lesser performances due to degradation phenomena that occur during the product’s first life and reprocessing [6~12]. Thus, the effect of reprocessing became a popular topic, and many studies have investigated the effect of reprocessing on their structure, thermal and mechanical properties [4, 13~17]. González-González, et al. [14] have identified the chain scissions during multiple extrusion which are linked to the melt temperature and resulted molecular weight losses. Brennan, et al. [18] recovered acrylonitrile-butadiene-styrene (ABS) and high-impact polystyrene (HIPS) from waste computer equipments, and found elongation at break and impact strengths were reduced considerably during reprocessing. Su, et al. [15] investigated the influence of reprocessing on the mechanical properties and structure of polyamide 6, and reported an increment in the tensile yield stress, flexural strength and modulus but a decrement in Izod impact strength and the molecular weight. Saffaro, et al. [4] found the tensile, flexural and impact properties of ABS deteriorated with reprocessing.

Polypropylene (PP), as one of the most common plastic materials, is usually being used in various applications while mixing with all sorts of fillers, such as minerals [19~23], clays [24] and fibres [25]. Among these fillers, talc is one of the most commonly used mineral filler which improves both the thermal and mechanical properties of PP [23], and a few studies have been conducted on reprocessing of talc-filled PP. Guerrica-Echevarria, et al. [13] studied talc-filled PP undergone multiple injection moulding cycles, and found that molecular weight decreased and break properties (such as Elongation at break) are related to the filler content and processing conditions. Bahlouli, et al. [26] studied the recycling effects on two high impact polypropylene (HiPP), and found a better thermal and structural stability for talc filled HiPP. Wang, et al. [23] pointed out that during the repeated extrusion cycles, talc in PP slightly increased the Young's modulus and the yield strength.

Yet, most current studies only focused on reprocessing effect on virgin composites, such as talc-filled virgin PP, or recycling of waste plastics from different sources. To the best of our knowledge, the influence of the filler content and reprocessing cycle numbers on the properties of recycled PP-based composites has received little to no attention. Lack of research on recycled plastic based materials would certainly limit their use, and it is both uneconomic and environment-unfriendly. Further, there is no research which focused on comparison of the effects of two major reprocessing routes (extrusion and injection moulding) on composites has been reported, and in industrial context, mechanical recycling usually involves both of the two procedures [27, 28]. In this paper, with the aim of promoting the understanding and use of composites based on recycled plastics, the effects from both extrusion and injection moulding cycles on the properties of talc-filled recycled PP composites were investigated via experimental methods. The results were discussed and correlated with previous researches.
which would provide a comprehensible knowledge base for reusing recycled plastics and their composites.

II. EXPERIMENTAL

A. Materials

The virgin PP (VPP) material used was a block co-polymer mainly being used for manufacturing automobile parts, with a trade-name of PPB-MO2-V and produced by Yangzi Sinopec. The material was used as received, and has an average particle size of 3.0 mm, density of 0.9 g cm\(^{-3}\), as shown in Figure 1. The recycled PP (RPP) used was some grey pellet recovered from white post-customer storage boxes, has an average particle size of 3.0 mm, density of 1.0 g cm\(^{-3}\), as shown in Figure 2, and was used as received.

The talcum powder (talc) used in this work was bought from a local factory, has an average particle size of 12.5 \(\mu m\), a density of 2.7 g cm\(^{-3}\), as shown in Figure 3, and was used as received.

The coupling agent, maleic anhydride grafted polypropylene (MAPP) used in this work was bought from Nanjing Deba Chemical Co., Ltd, has an average particle size of 2.5 mm, a density of 0.9 g cm\(^{-3}\), with the grafted rate of 0.8%, as shown in Figure 4, and was used as received.

The compositions used in the paper were shown in Table 1.

<table>
<thead>
<tr>
<th>Designation</th>
<th>PPB-MO-V</th>
<th>Grey RPP</th>
<th>Talc</th>
<th>MAPP</th>
</tr>
</thead>
<tbody>
<tr>
<td>VPP</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>RPP</td>
<td>0</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>RPP-T20</td>
<td>0</td>
<td>75</td>
<td>20</td>
<td>5</td>
</tr>
<tr>
<td>RPP-T40</td>
<td>0</td>
<td>50</td>
<td>40</td>
<td>10</td>
</tr>
</tbody>
</table>

B. Sampling and Reprocessing

1) Extrusion cycles

All materials for multiple extrusion cycles were processed by a Kangrun KRSHJ-20 extruder; a co-rotating, intermeshing twin-screw extruder, with screw diameter of 22 mm and \(L/D=44\). A single-screw feeder attached to the hopper was used for all the PP pellets. The processing temperatures were allowed to increase from 180°C to 200°C going from the hopper to the third barrel, and the temperatures of the last three barrels remained at 200°C, while the die temperature was set at 200°C. The screw rotation speed was 180 rpm, and the total mass flow rate was 5 kg h\(^{-1}\). Blended strands were extruded into a water bath for cooling, and then pelletized by a cutter. The average extruded pellet size was 2.8 mm. This process was repeated 5 times under the same operating conditions, so the grinded material of each cycle was the starting material for the following reprocessing cycles.

Some pellets were taken in every extrusion cycle for making testing sample pieces. Those taken pellets were dried in a dry oven at 85°C for 12 h with constant air flow to keep the moisture content below 1% before being fed into the injection moulding machine.

Then these were injection moulded into ISO standard test specimens using a Haitian MA1200/370 injection moulding machine, and 2 (tensile pieces) or 4 (flexural or impact pieces) test specimens were produced per single injection moulding process, see Figure 5. The temperatures of five heating barrels were set at 190°C, 192°C, 195°C, 200°C, 200°C, with injection pressure of 50 MPa, injection speed of 50 g per second, packing pressure of 30 MPa for 10 s, cooling in moulds was allowed for 10 s. The mould was pre-heated to 50°C. The
processing parameters were set in accordance with real manufacture [27].

2) Injection moulding cycles

All materials for multiple injection moulding cycles were processed by a Kangrun KRSHJ-20 extruder; a co-rotating, intermeshing twin-screw extruder, with screw diameter of 22 mm and L/D=44. A single-screw feeder attached to the hopper was used for all the PP pellets. The processing temperatures were allowed to increase from 180°C to 200°C going from the hopper to the third barrel, and the temperatures of the last three barrels remained at 200°C, while the die temperature was set at 200°C. The screw rotation speed was 180 rpm, and the total mass flow rate was 5 kg h⁻¹. Blended strands were extruded into a water bath for cooling, and then pelletized by a cutter. The average extruded pellet size was 2.8 mm.

The extruded pellets were dried in a dry oven at 85°C for 12 h with constant air flow to keep the moisture content below 1% before being fed into the injection moulding machine.

Then these were injection moulded into ISO standard test specimens using a Haitian MA1200/370 injection moulding machine, and 2 (tensile pieces) or 4 (flexural or impact pieces) test specimens were produced per single injection moulding process, the same as that shown in Figure 5. The temperatures of five heating barrels were set at 190°C, 192°C, 195°C, 200°C, 200°C, with injection pressure of 50 MPa, injection speed of 50 g per second, packing pressure of 30 MPa for 10 s, Cooling in moulds was allowed for 10 s. The mould was pre-heated to 50°C. The processing parameters were set in accordance with real manufacture [27].

This process was repeated 5 times under the same operating conditions. In each injection cycle, parts of the specimens were used for characterization purposes, while the remainders were grinded by a cutting mill (Retsch, Germany, model SM 200, as shown in Figure 6 left) in order to be reprocessed. The 4 mm sieve used was shown in Figure 6 right. The shredded plastic pellets were shown in Figure 7, with an average particle size of 3 mm.

C. Sampling and Reprocessing

1) Mechanical

The mechanical properties of composites were evaluated in terms of tensile, flexural, and impact properties, and all sample specimens were conditioned at 23°C and 50% R.H. for over 88 h before testing in accordance with ISO291 specifications [30].

The tensile properties tested were tensile strength, yield strength and elongation at break, assessed in accordance with ISO527 specifications [31]. The gauge distance is 110 mm with fixers' moving speed of 50 mm min⁻¹, sampling rate at 200 pts s⁻¹, full-scale load range of 20 kN, performed on a Gotech Universal Testing Machine (model TCS-2000NE) at room temperature of 23°C and at 50% R.H. The tensile modulus was obtained by an extensometer (Epsilon Technology Co.Ltd) at tensile elongation of 1%.

The flexural properties tested were flexural modulus and flexural strength, assessed in accordance with ISO178 specifications [32]. The span was set to 64mm at a crosshead
speed of 2 mm min⁻¹, sampling rate at 200 pts s⁻¹, full-scale load range of 20kN, performed on a Gotech Universal Testing Machine (model TCS-2000NE) at a room temperature of 23°C and at 50% R.H.

8 sample pieces were tested for each property tested and the average result taken if the coefficient of variance met the required limits (5% in accordance with ISO2602 specifications [33]).

2) Rheological

The flow behaviour of the materials was assessed using steady and dynamic shear rheology. The test was performed by utilising a dual-bore capillary rheometer (Rosand RH2200, Malvern Instruments) with two capillary dies with same radius of 1 mm but different length/radius ratios. Samples were pre-heated in the dual barrels at 190°C for 2 min, and measurements were carried out at 190°C under a shear rate ranging from 10 to 5000 s⁻¹, at room temperature of 23°C and at 50% R.H. Viscosity is plotted against shear rate, and the power law model was used to describe relationship between viscosity and shear rate as described in Eq.(1):

\[ \eta = K \gamma^{n-1} \] (1)

where the consistency K corresponds to the viscosity value for a shear rate \( \gamma \) of 1 s⁻¹ and the power-law index \( n \) characterizes the deviation of the Newtonian behaviour.

3) Thermal

The thermal properties of the materials were measured in form of the temperatures of deflection under load (TDL). The tests were conducted by using an HDT-VICAT test processor (CEAST model 6911.000) according to ISO75 [29], with constant heating rate of 50°C h⁻¹ and a load of 0.45 MPa. The samples were immersed in silicon oil which filled the tank and preheated for 4 min at 40°C, therefore the tests were carried out at a room temperature of 23°C and at 50% R.H. An average 6 samples were prepared and tested in each group.

III. RESULTS AND DISCUSSION

In this session, the following equation was used to calculate the degradation rate (DR) of the composites' performance which was applied for all properties:

\[ DR = \frac{P_o - P_a}{P_o} \times 100\% \] (2)

in which, the symbol \( P_o \) denotes the original performance obtained from initial tests once the specimens were made, the symbol \( P_a \) denotes the performance obtained after the ageing procedures were performed.

A. Effect of Reprocessing

Some of the experimental results from repeated extrusion cycles set were plotted in figures below.
From the figures shown above, the performance of the virgin PP decreased slowly with the number of extrusion cycles, except for elongation at break. This probably resulted from chain-scissions in polymers occurred in elevated temperatures and rotating shear forces [8, 9, 14].

For recycled PP and talc-filled recycled composites, some properties exhibited a little improvement with reprocessing cycles initially and decreased with further extrusion, such as yield strength and TDL, while other properties are shown to be deteriorated shown to be slightly with reprocessing cycles. The elongation at break and flexural strength of recycled PP remained stable, and were slightly increased at the 5th cycle. For talc-filled recycled PP composites, their performances were comparatively more stable during repeated extrusions except for elongation at break. The talc content improved the mechanical and thermal properties of recycled PP as predicted [20, 23].

DRs of some properties during extrusion cycles were summarized in Table 2 using Eq.(2).

**TABLE II.** **DRs OF SOME PROPERTIES DURING EXTRUSION CYCLES (%,** **COMPARING WITH THE 1ST PROCESS**.)

<table>
<thead>
<tr>
<th>Number of Cycles</th>
<th>VPP</th>
<th>RPP</th>
<th>RPP-T20</th>
<th>RPP-T40</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile Modulus</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>1.42</td>
<td>2.49</td>
<td>-0.33</td>
<td>1.85</td>
</tr>
<tr>
<td>3rd</td>
<td>4.89</td>
<td>6.50</td>
<td>-0.54</td>
<td>3.43</td>
</tr>
<tr>
<td>4th</td>
<td>7.54</td>
<td>7.79</td>
<td>-1.23</td>
<td>2.14</td>
</tr>
<tr>
<td>5th</td>
<td>8.53</td>
<td>12.86</td>
<td>0.48</td>
<td>3.31</td>
</tr>
<tr>
<td>Tensile Strength</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>-0.28</td>
<td>1.06</td>
<td>-0.06</td>
<td>-1.21</td>
</tr>
<tr>
<td>3rd</td>
<td>0.58</td>
<td>1.94</td>
<td>-0.07</td>
<td>-0.15</td>
</tr>
<tr>
<td>4th</td>
<td>0.06</td>
<td>1.51</td>
<td>-0.60</td>
<td>0.43</td>
</tr>
<tr>
<td>5th</td>
<td>1.07</td>
<td>2.29</td>
<td>0.63</td>
<td>0.00</td>
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<tr>
<td>Yield Strength</td>
<td></td>
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<td></td>
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<tr>
<td>2nd</td>
<td>0.50</td>
<td>0.30</td>
<td>-0.79</td>
<td>-0.95</td>
</tr>
<tr>
<td>3rd</td>
<td>1.19</td>
<td>0.07</td>
<td>-0.87</td>
<td>-1.44</td>
</tr>
<tr>
<td>4th</td>
<td>0.36</td>
<td>-0.91</td>
<td>-2.84</td>
<td>0.51</td>
</tr>
<tr>
<td>5th</td>
<td>3.73</td>
<td>3.49</td>
<td>0.12</td>
<td>1.54</td>
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<tr>
<td>Flexural Modulus</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>3.95</td>
<td>0.50</td>
<td>0.12</td>
<td>1.54</td>
</tr>
<tr>
<td>3rd</td>
<td>4.37</td>
<td>2.35</td>
<td>2.65</td>
<td>1.07</td>
</tr>
<tr>
<td>4th</td>
<td>4.87</td>
<td>4.79</td>
<td>2.76</td>
<td>1.94</td>
</tr>
<tr>
<td>5th</td>
<td>6.04</td>
<td>6.03</td>
<td>3.13</td>
<td>2.55</td>
</tr>
<tr>
<td>Flexural Strength</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>0.62</td>
<td>0.89</td>
<td>-1.24</td>
<td>1.68</td>
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<tr>
<td>3rd</td>
<td>2.01</td>
<td>2.57</td>
<td>0.67</td>
<td>1.44</td>
</tr>
<tr>
<td>4th</td>
<td>2.14</td>
<td>1.40</td>
<td>-1.23</td>
<td>1.39</td>
</tr>
<tr>
<td>5th</td>
<td>2.92</td>
<td>0.14</td>
<td>-0.57</td>
<td>1.96</td>
</tr>
<tr>
<td>TDL</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>2.00</td>
<td>-0.13</td>
<td>-1.19</td>
<td>-1.56</td>
</tr>
<tr>
<td>3rd</td>
<td>3.02</td>
<td>-5.28</td>
<td>0.87</td>
<td>-0.42</td>
</tr>
<tr>
<td>4th</td>
<td>3.65</td>
<td>-2.07</td>
<td>2.81</td>
<td>-0.10</td>
</tr>
<tr>
<td>5th</td>
<td>4.74</td>
<td>1.61</td>
<td>1.95</td>
<td>1.25</td>
</tr>
<tr>
<td>Average</td>
<td>2.91</td>
<td>2.17</td>
<td>0.37</td>
<td>0.94</td>
</tr>
</tbody>
</table>

In Table 2, virgin PP showed the largest DR values, because at low molecular weight (MW), the chain scission is random, but at higher MW it becomes MW-dependent, increasing with MW [35], and MW of virgin PP is larger than recycled PP [36].

In repeated heat-and-shear-involving cycles, the polymeric chain lengths become more unified than original lengths and MW, as shown in other’s study [35], and the amorphous phase between the lamellae that has an increased mobility is increasing with the number of reprocesses [22], it could gave those polymers a better elongation at break. On the other hand, the presence of talc and contamination within PP matrix resulted in an increase in crystallinity [19, 23, 37], which would increase tensile properties, and reduce elongation at break. Thus, it is highly complicated in fully understand the pattern of recycled PP/talc composites, as shown in figures and Table 2, and needs further study. Still, the stabilizing effect of talc content was observed in recycled composites, as it did same effect on reprocessing of virgin PP/talc composites [23, 26, 37].

For rheological behaviours in the form of shear viscosity plot of the composites shown in Figure 15, and it quite obvious that reprocesses decreased the shear viscosity of virgin material, which might contribute to chain-scission actions took place in elevated temperatures during reprocessing. However, some talc-filled recycled composite
(RPP-T20) was comparatively more stable during multiple heat-involving procedures, as shown in Figure 15.

The results from repeated injection moulding cycles have shown the same pattern. It is supposed that the presence of talc does not cast a significant influence on the degradation mechanisms of the material matrix [13]), and the stability of recycled PP/talc composites compared might due to a gradual increase of delamination and dispersion of talc particles or agglomerates during the successive reprocessing, which resulted in an increased number of particles and a decreased particle size [23]. The consistency of flow properties of recycled PP/talc composites could lead to a more stable production rate, which would facilitate the use of such composites.

B. Comparison of Effect on Recycled Composites

Tensile results of talc-filled recycled composites from repeated injection moulding cycles set were plotted in Figure 16 to Figure 18, with comparison with multiple extrusion test set, since the tensile properties were found to be critical properties in reprocessing study [34].

As shown in those figures, the materials processed by multiple injection moulding cycles are still preserving some good characteristics as the same as those being processed by multiple extrusion cycles. The tensile strength and yield strength are increased with number of injection moulding cycles, which would imply the increase of crystallinity as mentioned before. All tensile properties of RPP-T40 were increased initially, and then decreased with further re-injection moulding, for it is possible that crystallinity degree of RPP-T40 has reached certain limit, and the amorphous phase increased with further reprocessing.

For the injection moulding cycles involved shredding procedure, which also could be considered as an elevated temperature and high shear rate procedure, the materials endure twice such processes when compared to extrusion set. The chain-scission effect took the ruling place in material matrix, and it would explain the lower tensile modulus than extrusion set.

To sum up, the recycled PP/talc composites still maintain some good performance during reprocesses, and were more stable when comparing to both virgin and recycled PP in most properties, as shown in the figures Table.2. Thus, reusing of rejected parts which made from the talc filled recycled composites could be feasible.
The purpose of this paper is to investigate the effect of reprocessing on recycled material, for fulfilling the knowledge gap resulted from lack of such research and promoting the use of recycled plastic, and using recycled plastics has been already proved to be both economic and environment-friendly.

For the first time, recycled PP was compounding with talc and coupling agent with different concentration for reprocessability study, where virgin and recycled PP were used as comparatives. The materials were subjected to 5 reprocessing cycles from two different routes, extrusion and injection moulding. The experimental part consists of mechanical, rheological and thermal.

The results showed the talc content has similar effect as it did in virgin PP, such as performance enhancing and stabilization. The recycled PP/talc composites were superior to virgin PP in most properties, and were more stable under multiple reprocessing cycles than both virgin and recycled PP. In both reprocessing routes, the performances of the recycled PP/talc composites were similar and constant, it showed the potential of using recycled materials.

There are still something unclear in this moment which displayed in complicated pattern of some properties. It requires further study to understand the competing mechanisms within recycled polymer composites under reprocessing, and how talc stabilize the recycled material.

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