Durability of Repetitive Polypropylene Recycling: Challenge on Securing The Mechanical Properties

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Abstract. Most of the products used in our daily lives involve plastic. However, plastic pollution can cause an incalculable environmental crisis. Hence, an extensive effort has been made towards recycling plastic since the last decade. The challenge on plastic recycling is to secure the mechanical performance of the products. In this study, the durability of repetitive polypropylene processing from ten to fourteen cycles is investigated. Optimum parameters such as moulding temperature, injection pressure, holding pressure, holding speed, holding time, and cooling time were used to successfully manufacture extensive recycled polypropylene by injection molding process. The tensile and bending strength of polypropylene were managed to maintain above 30 MPa even at fourteenth cycles. The bending modulus was secured at least 1.0 GPa for the polypropylene after recycled for fourteen times. Besides, the recycled polypropylene could sustain more loading in bending mode rather than longitudinal tension tight. The percentage of shrinkage was less than 3%. This little shrinkage gave an assurance to the dimensional stability even though the reprocessing was at high level until fourteenth cycle. Scanning electron microscopy (SEM) images evident the homogeneous mixing and brittle manner of polypropylene during extensive recycling.

1. Introduction
Today’s plastic production and demand around the globe are rapidly increasing to an extent that it needed to be taken care of. This is because most of the products we used in our daily life technically are from plastic. Plastics are durable, lightweight and cheap compared to other conventional materials such as steel and ceramics. They can be readily molded into various products which can suit many applications. Unfortunately, million tons of plastic used every day are causing the pollution in the environment and is giving a huge negative impact to the earth [1]. This is because plastic takes a long time for decomposition. Energy intensive techniques of plastic manufacturing method may destroy a fragile ecosystem. It gives serious effect to marine environment, food chain and the groundwater because of the toxic chemical reaction during incineration. Hence, plastic waste becomes a source of global contamination that will continue to the future.

Fortunately, some plastics are suitable for recycling but their waste is still abundant in the world. Consequently, the reuse, recovery, and recycling of plastics becomes waste management strategy. There is a report made by the Environmental Protection Agency based on accumulated data until 2015
In the report, surprisingly, it showed that there was only 7% out of several tons of plastic wastes were recycled. Thus, recycling processes are encouraged as the number of contaminants are increasing. This effort draws attention from the world community that relates to environmental preservation. Recycling of plastics refers to the process of reprocessing the waste or scrap plastic materials into useful products. This approach aims to bring down the rates of plastic pollution. Furthermore, recycled plastics are lower in price. On the other hand, recycling system give positive environmental benefits in terms of energy recovery [3]. One of the methods to recycle the plastic is using plastic injection molding. This method can fabricate the products in a high production rate. These benefits contribute to world economy and industrial ecology.

Nowadays, polypropylene is one of the famous materials especially in the making of the personnel protective equipment (PPE) during pandemic. The polypropylene also involved in fabricating food packaging, equipment manufacturing and electrical component. Not only that, automotive industry uses polypropylene. Automotive industry uses polypropylene for car bumpers, battery casings and interior parts [4].

A lot of studies proved that thermoplastic can be reused or recycled, but none of them can provide the exact limit of recycling [5-7]. Bahlouli et al. investigated the number of recycling process up to 12 cycles. The process led to the decrease of the molecular weight, failure stress and impact energy [8]. In another study, the appearance of the product was changed and became fragile when number of recycling process was increased until five cycles [9]. There is a report revealed that the recycled polypropylene was good performed in tension but not to impact [7].

In fact, the recycling process became complicated in production lines if the recycled plastic is incorporated with additives or blends with other types of plastic. A major challenge may be faced when the plastic wastes constitute of different types, which are inherently not compatible to each other. This problem will increase the difficulty of recycling. The most economic and viable production is a direct recycling of mono-type polymer. Thus, this study focused on the repetitive production of homo-polypropylene from ten until fourteen times recycling. The challenge on securing the tensile and bending performance of recycled polypropylene is studied. Besides, the dimension stability as a crucial factor in product design is also investigated.

2. Recycling process and testing
The polypropylene was mechanically recycled in this study. The raw materials of this study are polypropylene (PP) homo-polymer grade PM200 with melt flow rate (MFI) of 16 g/10 min. This type of PP is suitable for extrusion, injection moulding or other conversion process. The materials were supplied from Lotte Chemical Titan (M) Sdn. Bhd. A granulator was used to produce pellets resin by crashing the product into smaller and fine granules. The average diameter of the resins was less than 4 mm. The model of the granulator was ML-SC5-150 and the machine was made from Ming Lee Industrial, Hong Kong. The granulator was cleaned before proceeding to the next cycle to avoid any contamination.

The repetitive recycling process was carried out using a MA23 model injection molding machine. The recycling process was continued up until fourteen cycles. Thus, optimum parameters of injection molding process must be set to ensure the successful manufacture at high level of reprocessing. We have reported the optimum parameters using Taguchi analysis in previous work [10]. The molding temperature was set at 200 °C. The injection and holding pressure were constant at 90 bar and 70 bar, respectively. Both the holding and cooling time were set at 6 seconds. The injection speed was at 40 rpm. A defect free specimen was successfully manufactured as shown in Figure 1.
The recycled polypropylene specimens were investigated by applying tensile test according to ASTM D638. The ASTM D-638 specifies the standard dimension of the tensile specimen of plastic and outlines accurate requirements for the test. The dimension of the specimen was determined using Type I as stated in ASTM D-638. The machine used for this tensile test was SHIMADZU AG-IS 50KN tensile machine. The test was conducted at the cross-head speed of 5 mm/min. The applied load was 10 kN to ensure the specimen failure at break. The average result was obtained from 5 measurement for each cycle.

Molding shrinkage ratio is one of the most important factors in the design of plastic injection molding. The shrinkage must as low as possible to maintain the dimension of final product as required. Thus, the percentage of shrinkage volume ratio of final product was determined using formula as shown in equation (1) [10]:

$$S(\%) = \frac{(V_i - V_o)}{V_o} \times 100$$

where $S$ is the percentage of shrinkage, $V_o$ is the volume of the product's mold cavity and $V_i$ is the volume of the final product. A digital vernier caliper with accuracy up to 0.01 mm was used to measure the dimension of the specimen.

A three-point bending test was conducted on the recycled specimens to determine the bending strength and bending modulus. The dimension of the specimen was according to ASTM D790. The experiment ran at the same speed as in the tensile testing using the same machine. The bending test was stopped after achieved 5% deflection.

The fracture surface of the specimen was examined after the Tensile test using Scanning Electron Microscope (SEM) model Hitachi TM3030Plus. The type of fracture was then determined.

3. Results and discussion

Table 1 summarizes the tensile results for recycled polypropylene from ten to fourteen recycling. The maximum tensile strength happened near the yield point because the failure had no or little necking. The polypropylene managed to sustain the tensile strength at fourteenth cycle. Tensile strength is not reduced significantly when the number of recycling was increased. The elastic modules from tenth to fourteenth recycling of polypropylene is in the range of 806 MPa to 865 MPa. The difference is only about 2%. However, the elasticity is not as good as the virgin polypropylene having above 1.0 GPa Young’s modulus [11]. It is noted that plastic recycling has suffered the chain scission effect due to thermal stress. During chain cleavage, de-polymerization weakens the capacity of the plastic to withstand the external force thus more easily deformed.

Strain is the deformation of the specimen. It relates with the ductility of the material. As PP is a brittle material, therefore the strain is small. The strain is less than 11% in this study which is in the
acceptable limit. This is because the bonding of the molecular chain is strong and can hold them together for a long time before it breaks [12].

| Number of recycling | Tensile strength at yield, $\sigma$ (MPa) | Elastic modulus, $E$ (MPa) | Maximum strain, $\varepsilon$ (%) |
|---------------------|------------------------------------------|---------------------------|----------------------------------|
| 10                  | 31.67±0.46$^a$                          | 806.16±14.89              | 10.05±0.26                       |
| 11                  | 32.35±0.31                              | 865.41±33.34              | 9.59±0.57                        |
| 12                  | 32.62±0.34                              | 832.73±32.69              | 9.93±0.15                        |
| 13                  | 32.97±0.37                              | 835.72±95.91              | 10.05±0.60                       |
| 14                  | 32.63±0.42                              | 841.87±64.37              | 9.46±0.33                        |

$^a$ standard deviation calculated based on the confidence level of 95%.

A comparison on recycling potentials among the famous plastics such as high-density polyethylene (HDPE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC) was studied [13]. It was found that PP has the highest recycling potential after considering the characteristics of recycled plastics. Hence, a little drop of tensile performance at fourteenth cycle still can be rescued by mixing with the pure polymer itself [7, 11].

Figure 2 displays the bending strength at 5% maximum deflection and bending modulus of the polypropylene from ten to fourteen cycles. It is noticed that the bending strength is higher than the tensile strength. The specimen produced at eleventh cycle exhibited the highest bending strength and modulus. Anyway, the bending modulus of all the recycled polypropylene is above 1.0 GPa. It is seen that bending modulus is much higher that Young’s modulus. This shows that the PP can sustain more loading in bending mode rather than longitudinal tension tight.

Both tensile and bending tests applied forces to the specimen. For a tensile test, the maximum stress is experienced throughout the entire volume. Nevertheless, the maximum stresses are conversely concentrated in a small region on the top surface of the specimen in the bending condition. Thus, the deformation is more serious at the tensile mode where a large area was affected. Besides, the semi crystalline behaviour of polypropylene also makes it tough but flexible with the ability to bend without breaking [11].
Figure 2. Bending properties of recycled polypropylene (a) strength (b) modulus

Figure 3. Shrinkage of recycled polypropylene at different number of recycling

The shrinkage percentage of recycled polypropylene is decreased as the number of recycling increases, as shown in Figure 3. Polypropylene is categorized in the group of semi-crystalline polymer. The shrinkage ability is pronounced if compared to amorphous type. Semi-crystalline polymer will rearrange their molecular chains to form crystalline during cooling process that lead to the less space needed for the same number of atoms [11]. As overall, the shrinkage for all specimen is less than 3%. Moreover, the recycled specimen has less than 1% at fourteenth cycle. The least shrinkage on the product is a warranty to the manufacturer particularly for good dimensional stability and accuracy.

Figure 4 shows a specimen recycled at fourteenth cycle that was broken at the middle of the specimen after tensile test. The specimen shows a brittle manner in failure. This is evident with the flat surface of fracture. The specimen did not reveal the cup and cone fracture that shows the significant
trend of ductile likes material. SEM image shows the tensile force pulling effect on the fracture surface in Figure 4(b). It is revealed that the longitudinal direction of stretch lines as a result of tensile force. The flat surface without necking is clearly seen in the SEM image evident no or little deformation in the brittle polypropylene.

![Figure 4. Fracture specimen (a) brittle mode (b) SEM image at fracture surface](image)

4. Conclusion
This study provides an evaluation on the durability of polypropylene recycling. The recycled of polypropylene was mechanically recycled until fourteen cycles without any defect. Although the polypropylene was repetitively ground, re-melted and re-fabricated, the tensile and bending performance was not seriously deteriorated after fourteen recycling. The tensile and bending strength was performed closed to virgin polypropylene, where the value was above 30 MPa. The elastic modulus was slightly lowered but the bending modulus achieved as high as 1.4 GPa. The challenge on securing mechanical performance of the polypropylene especially on bending properties was a success. The promising result was the stability in dimensional control where the shrinkage was less than 3%. The findings from this study conclude a high potential recycling for polypropylene until the fourteenth cycle. This study can contribute substantially to the manufacturing industry in waste management of plastic products. The cost and the use of plastic materials can be reduced to make the environment to be more protected through energy recovery. This study also gives an improved basic on the recycling the polypropylene waste in the injection molding industries.

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