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Unveiling Polypropylene's Repetitive Recycling Potential at High Temperatures

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ABSTRACT

Polypropylene (PP) is renowned for its lightweight, chemical stability, hygienic properties, and low cost, making it a valuable material in many applications. However, improper disposal of PP presents significant environmental hazards. One of the efforts made to reduce plastic pollution is through recycling. The semi-crystalline behavior of PP is prone to degradation in its mechanical performance, especially when the plastic undergoes a high-temperature reheating process. This study investigated the degradation limits of recycled PP produced from injection molding from the 15th to 19th cycles at two different temperatures, namely 220 °C and 240 °C. A comparison of the tensile and bending properties of the recycled product was made for these two temperatures to determine the maximum limit of PP degradation. When recycling began at the 15th cycle, there was a significant reduction in tensile and bending properties compared to the original PP. Subsequently, the degradation rate was relatively slow from the 15th to the 19th cycles. As the recycling temperature increased, it was found that the degradation of yield strength became more severe. The decrease in bending strength could be more pronounced with the effect of temperature. The color transformation of recycled PP, resulting from the chain scission effect, contributed to the deterioration of its mechanical properties. Thus, industrial users must consider the down-cycling resilience to extend PP recycling beyond 15 cycles.

INTRODUCTION

Plastics are used in numerous applications, such as packaging, building and construction, textile, automotive, electrical, and electronic parts. There is still a growing global demand for plastics, with an expected increment rate of 5 % annually [1]. Amid the COVID-19 pandemic, plastic packaging, as a

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guarantor of good safety and hygiene, is emphasized [2] where many disposable medical equipment made of plastics, such as gloves, disposable syringes, facial masks, sterile packaging, and personal protective equipment (PPE). The widespread use of single-use plastic packaging and disposable medical equipment has significantly contributed to the escalating amount of plastic waste in landfills and oceans. It has been reported that more than 8 million tons of plastic are discarded in landfills and the environment every year [1]. The persistence of non-biodegradable plastic contributes to plastic pollution and poses a significant threat to global ecosystems. Thus, balancing the benefits of plastic with environmental and health challenges requires sustainable practices, responsible consumption, and ongoing efforts to develop alternative materials and recycling solutions. Currently, many countries worldwide are keen to find ways to maintain the convenience of plastic while minimizing its negative impacts on the environment and human health. Continuous research and development efforts are essential to effectively solve the global plastic waste crisis by adopting the 7R's sustainability model, which includes replacing, recycling, redesigning, reusing, reposting, remodifying, and recovering [3].

In this study, polypropylene (PP) is the key factor investigated due to its application as single-use plastic packaging products [4]. Mannheim and Simenfalvi [5] examined the environmental impact of PP products. They found that 91 % of the environmental load was contributed by the production stage of PP, particularly the injection molding process. PP recovered from waste is more promising than other plastics and has been proven to be effectively recovered through optical infrared separators [6]. Many studies [6], [7], [8] have reported on mixing recycled PP with other materials to sustain the mechanical performance of the final products. Recycled PP could be added with virgin PP [6] or other natural materials [7] to mitigate plastic pollution while increasing the economic value of PP waste.

The injection moulding process is the most popular manufacturing method used in the plastic industry due to its high production rate, design flexibility, product dimension, and quality consistency. The types of waste produced after the injection molding process are flash, sprue, runner, and rejected parts, which are then regarded as plastic waste. However, only 13 % of plastic waste was reused and recycled [9], [10]. Thus, this study investigated the feasibility of reusing, redesigning, and reproducing the PP products from PP waste using mechanical recycling. This study only focused on the degradation limit of PP recycling in the injection molding process without incorporating additives. PP's tensile and bending properties were also determined when exposed to repeated high temperatures during recycling. The findings of this study are important for promoting eco-friendly engineering solutions to the plastic industry.

EXPERIMENTAL WORKS

The raw material employed in this study was homo-polymer PP grade PM200 supplied by Lotte Chemical Titan (M) Sdn. Bhd. The raw material's melt flow rate (MFR), according to ASTM D1238, was 16 g/10 min. No additives were used in the injection molding and recycling process. Virgin PP samples were fabricated using an MA23 injection molding machine at two different molding temperatures, namely 220 °C and 240 °C. The injection temperature and time for sample fabrication are tabulated in Table 1.

Table 1. Injection moulding process parameters

Parameter and unit	Value
Injection pressure, MPa	140
Holding pressure, MPa	70
Holding time, s	6.0
Cooling time, s	10.0

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The PP samples and runner system are shown in Fig. 1. The PP samples and runners were crushed after the injection molding process. The first cycle of PP granules, with an average diameter of 1 mm to 2 mm, was produced using a low-speed granulator. The regrind obtained from the crushing process was injected, followed by repeated crushing until a 19-hour cycle using the same injection molding process parameters. Results from the 10^{th} to 14^{th} cycles have been reported [11], whereas this study focused only on the 15^{th} to 19^{th} cycles.

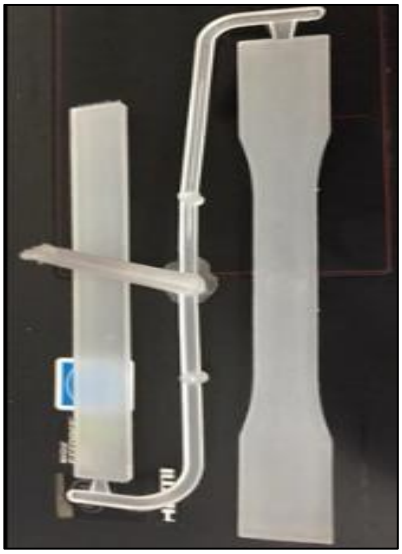

Fig. 1. Virgin PP samples and runner system.

Recycled samples were subjected to tensile and three-point bending tests according to the standard dimension of the sample and operating procedures stated in ASTM D-638 and ASTM D-790, respectively. Virgin PP samples were also tested as a reference. A dog bone-shaped sample was used for the tensile test, whereas a rectangular bar was used for the three-point bending test. Both tests were conducted using the MIMADZU AG-IS 50 kN universal tensile machine at a 5 mm/min crosshead speed. The mean value was determined from five measurements for each test. In a tensile test, the sample failed at break, while the bending test ceased when the sample achieved 5 % deflection without breaking. The tensile test sample's fracture surface was observed using Hitachi TM3030Plus scanning electron microscope (SEM). A flow chart of experimental work is presented in Fig. 2.

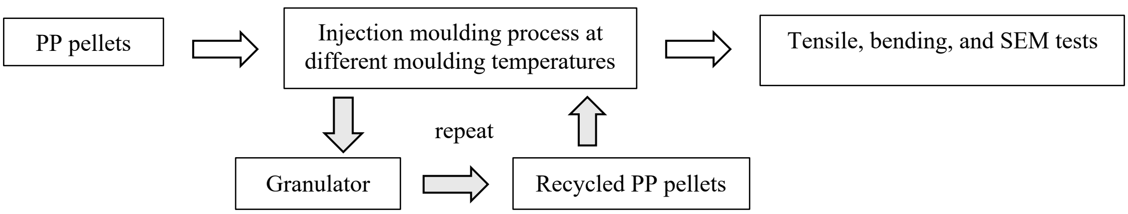

Fig. 2. Flow chart of experimental work.

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RESULTS AND DISCUSSION

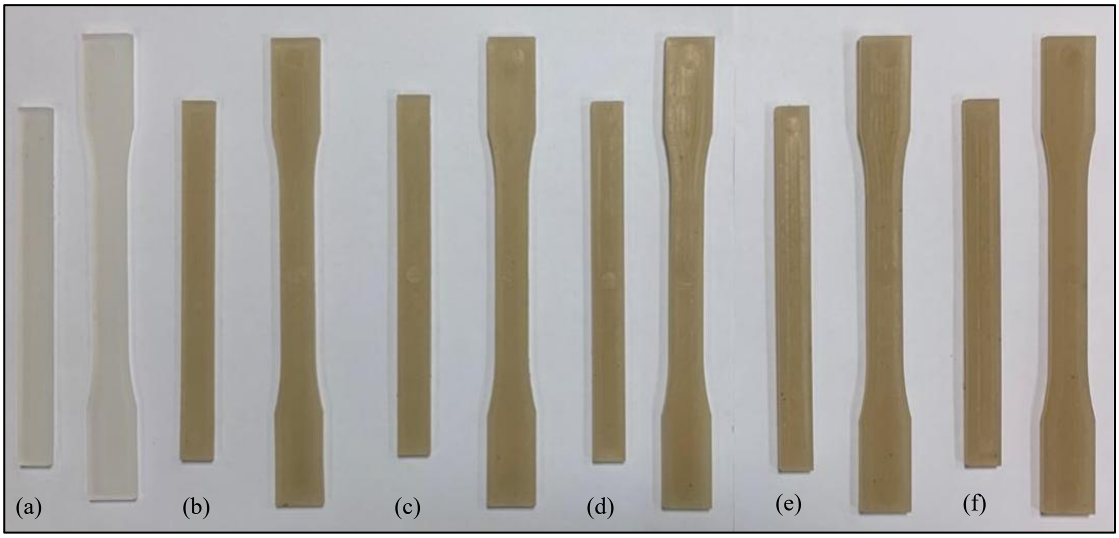

Fig. 3. Appearance of recycled PP at 240 °C (a) virgin (b) 15th cycle (c) 16th cycle (d) 17th cycle (e) 18th cycle (f) 19th cycle.

Fig. 3 and Fig. 4 display the difference in the appearance of recycled PP at different temperatures and cycles. The virgin PP was white and transparent. A significant transformation from white to brown is observed in Fig. 3 when the PP samples underwent repeated crushing and heating in the recycling process. The brownish color darkened with increasing temperature and reprocessing cycles. The color changes indicated changes in the molecular structure of PP. A similar finding is supported by another research [12].

The samples absorbed thermal stress during the polymerization process in the injection molding process. Absorption caused the structural bond between hydrogen and carbon atoms in the PP to become weaker. This resulted in the chain scission effect. The PP polymer was depolymerized to form free radicals and experienced thermal oxidization. The degradation mechanism of PP during recycling may alter PP's physical and mechanical properties. It can be seen in Fig. 4 (c) that the tensile specimen reflected a flat fracture without necking. The chain scission effect causes PP to form shorter polymer chains [12], making it more brittle.

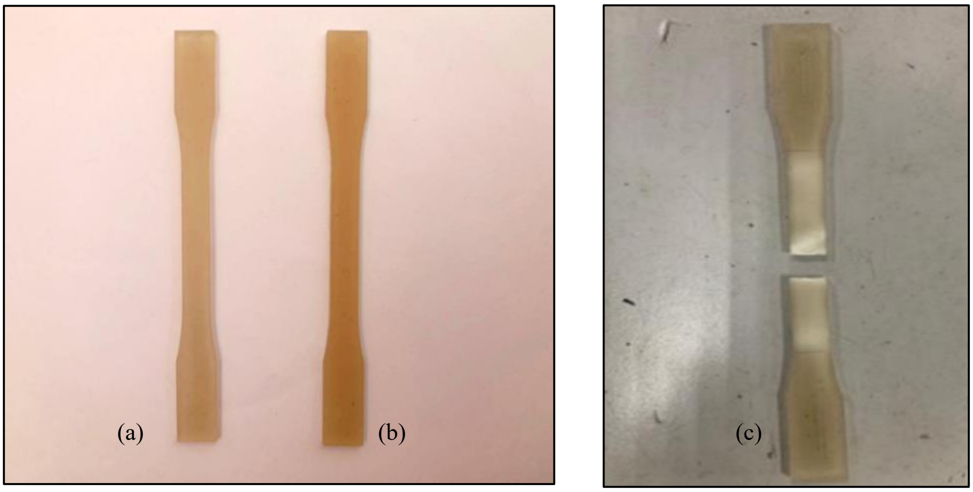

Fig. 4. Appearance of recycled PP at different conditions: (a) 220 °C, (b) 240 °C, and (c) fracture of tensile specimen for 15th cycle.

Fig. 5 presents the SEM images of the fractured specimen at the 19th cycle before and after undergoing the tensile test. Fig. 5 (a) shows the homogeneous surface of recycled PP without precipitates, which exhibited the smooth fluidity of recycled granules. It has been reported [13] that substantial shear forces destroy the molecular chains of PP during crushing and repeated heating. This phenomenon led to a decrease in the average molecular weight, an increase in the melt flow index, and an increase in the number of recycling cycles. Therefore, recycled PP granules were easily filled in the mold during injection when the material fluidity gradually increased. The longitudinal shear lines visible on the surface of PP after the tensile test, as shown in Fig. 5 (b), revealed the tensile force exerted on the sample. Fig. 5 (c) shows the cross-section of the fracture surface after the tensile test. A straight fracture appeared, indicating brittleness without necking or plastic deformation.

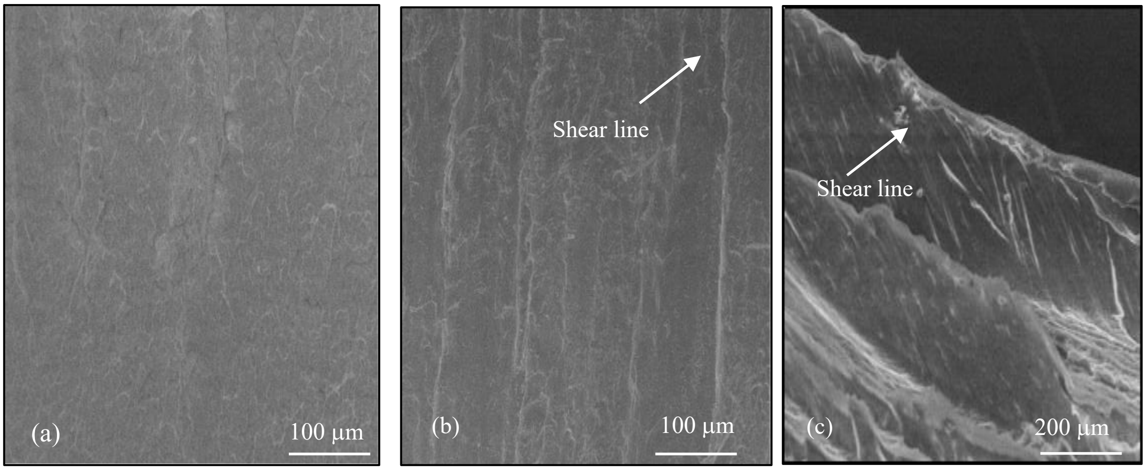

Fig. 5. SEM images of the specimen after the 15th cycle and temperature of 220 °C: (a) surface before the tensile test, (b) surface after tensile test, and (c) cross-section of the fracture surface.

From Fig. 6(a), it was found that the yield strength significantly declined to below 30 MPa at moulding temperature of 220 °C, and the yield strength decreased to below 29 MPa at moulding

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temperature of 240 °C. According to previous work [8], recycling up to eight times caused the mechanical properties of plastics to deteriorate by up to 20 %. in this study, the difference in yield strength of recycled pp ranged from 13 % to 18 % when compared to virgin pp (34.30 Mpa). Oliveira et al. [12] stated that the strength properties are strongly influenced by the chain scission effect promoted in the recycling process. evidence of colour change observed in the recycled pp indicated that chain scission took place in the amorphous regions, further reducing the stiffness of the recycled pp. However, no significant reduction trend was observed in yield strength and elastic modulus with increasing recycling cycles up to 19 times. The elastic modulus slightly increased when the moulding temperature was increased from 220 °C to 240 °C. It was discovered that the mold temperature had a significant impact on the Young's modulus, followed by barrel temperature. An increase mould temperature may increase Young's modulus due to changes in crystallinity. This similar finding is supported by Leyva-Porras et al. [14].

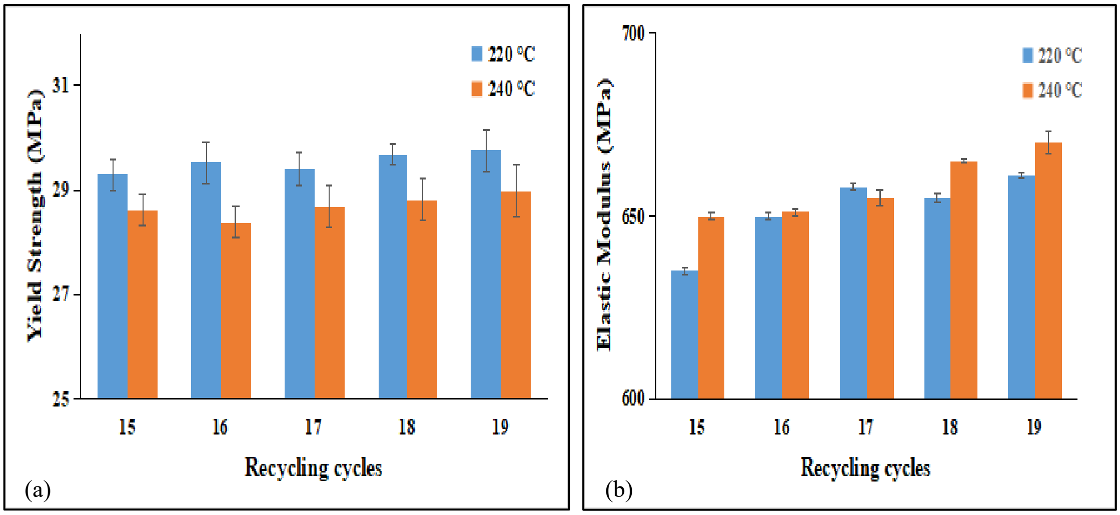

Fig. 6. Effect of recycling cycles and moulding temperature on (a) yield strength and (b) elastic modulus of PP.

The strain was consistent regardless of the effect of recycling cycles, as shown in Fig. 7. The maximum strain obtained by recycled PP was about 13 % at 220 °C but reached 11 % at 240 °C. Oblak et al. [15] reported similar findings, where the elastic modulus increased when the recycling process was extended to the 9th cycle. It may be due to chain scission and thermomechanical degradation effect. These effects loosened the molecular chains of pp [13] and disorganized them into smaller molecular chains. smaller polymer chains have less capability to resist external load [11] and break at lower elongation [15]. The elastic modulus of recycled PP at both temperatures was reduced by about 25% compared to the elastic modulus of virgin PP, which was more than 800 MPa.

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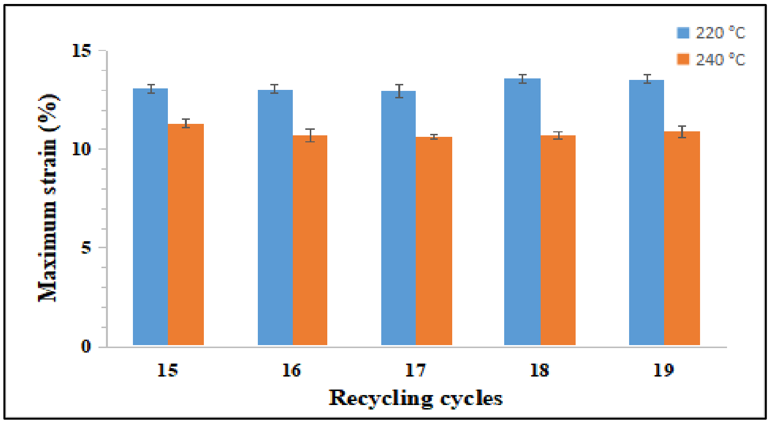

Fig. 7. Effect of recycling cycles and moulding temperature on maximum strain at break of PP.

Based on Fig. 8, the bending strength at 5% deflection exhibited a reduction trend compared to the original PP (40.05 MPa). Both bending strength and modulus at 240 °C were lower than at 220 °C. The difference in bending strength between recycled PP and virgin PP was achieved from a minimum of 20 % at 220 °C to 35% at 240 °C. Deterioration in bending properties was supported by Tesfaw et al. [16]. The bond strength became weaker in 100% recycled plastic when the plastic underwent various types of stresses during the recycling process, such as injection and crushing.

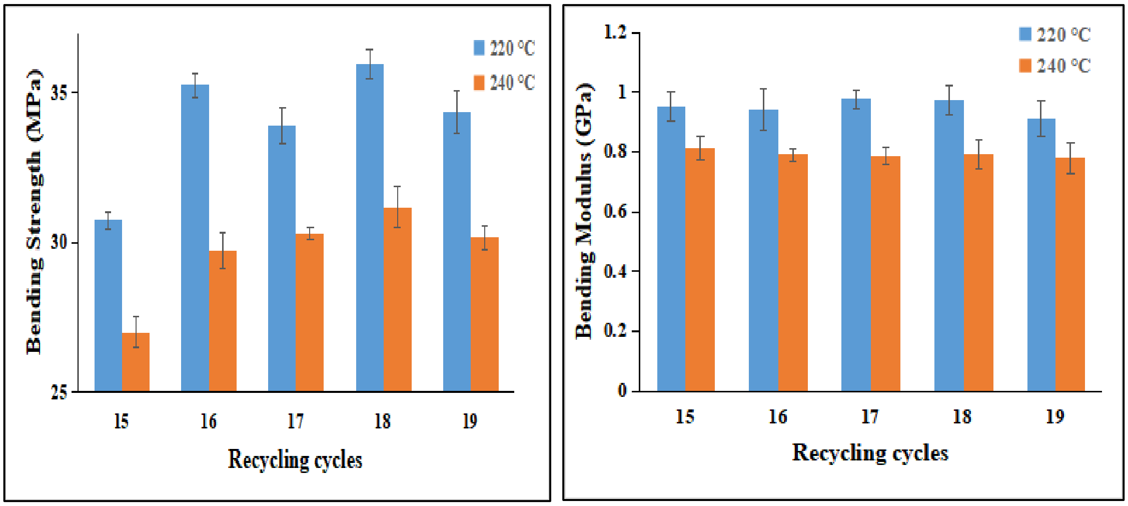

Fig. 8. Effect of recycling cycles and molding temperature on (a) bending strength and (b) bending modulus of PP.

The temperature effect was more pronounced in bending properties, which involved localized stress concentration. the difference in tensile strength between the two temperatures was less than 4 %, but the difference in bending strength between the two temperatures was more than 12 %. moreover, the bending modulus of recycled PP maintained almost 1 GPa at 220 °C, but the bending modulus of recycled PP decreased below 0.8 GPa when the reprocessing temperature increased to 240 °C. Therefore, higher temperatures may accelerate the thermal degradation of plastics and further reduce the deformation resistance under load.

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CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted without any self-benefits or commercial or financial conflicts and declare the absence of conflicting interests with the funders.

AUTHORS' CONTRIBUTIONS

Koay Mei Hyie and Normariah designed and supervised the research. Hamid and Suhada provided the theoretical framework. Mohd Hidayat Wadi carried out the research. Nor Azirah anchored the data analysis and review of the article submission.

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