Degradation Effects of Primary Recycling on Polypropylene in Injection Moulding Process

Alexandre Mateus Popiolek[a]

[a] Federal University of Santa Catarina, Florianopolis, Brazil, alexandreja_@hotmail.com.

Abstract. This review investigates the degradation effects of polypropylene (PP) during primary recycling and injection moulding (IM), focusing on its reprocess-ability and impact on material properties. Given PP's extensive use in various industries and its strategic role in sustainability efforts, understanding its recycling behaviour is essential for optimizing industrial practices and minimizing environmental impact. The study examines how repeated thermal and mechanical exposure influences PP's molecular structure, mechanical performance, and crystallization characteristics, aiming to provide a comprehensive assessment of its suitability for multiple recycling cycles. Chain scission is identified as the dominant degradation mechanism, contributing to a progressive decline in molecular weight, alterations in melt flow index, and modifications in crystallization behaviour. The evaluation includes a discussion of the effects of recycling on stiffness, yield strength, toughness, and viscosity, alongside changes in microstructural attributes such as spherulite formation and crystallization kinetics. By analysing processing parameters—including screw profile aggressiveness—and their role in shaping PP's degradation trajectory, this review seeks to establish guidelines for maintaining material integrity and ensuring performance consistency in recycled PP applications. The findings highlight the importance of optimizing recycling protocols to mitigate degradation-induced limitations, reinforcing the need for industry-driven improvements in polymer recovery strategies. Moreover, the study synthesizes findings from various studies to provide a comprehensive assessment of PP's degradation pathways, aiming to refine reprocessing strategies that mitigate material deterioration. Understanding these mechanisms is essential for optimizing recycling processes and improving the consistency of properties in recycled PP, ensuring its continued applicability in manufacturing while maintaining the integrity of IM products.

Keywords. Polypropylene, Primary recycling, Injection moulding, Material degradation, Reprocess-ability.

1. Introduction

Plastics are widely used in various industries due to their malleability and versatility [1] and have experienced an exponential increase from 1.5 to 390 million tons over the past decades (1950 to 2021) [2,3], with a projection of 460 million tons by 2030 [4]. With rising oil prices and the hypothesis that plastic products may last millennia before decomposing, depending on physical-environmental conditions and chemical composition, there is a growing demand for recycling, considered the best alternative as it reduces the demand for virgin materials and disposal [5-7].

Thermoplastic waste can be categorized into three groups according to ASTM D5033 [8]: primary recycling, which reprocesses clean plastic waste into products with similar properties; secondary recycling, which converts mixed or contaminated plastics into lower-quality materials; and tertiary recycling, which breaks down plastics into chemical components for reuse [9].

Primary recycling is both economically and environmentally advantageous and is easy to implement, as it bypasses the need for sorting, filtering, or cleaning operations typically required for materials from landfill sites or post-consumer waste [9, 10].

Table 1 shows the terminology of ASTM D5033, ISO 15270 and other equivalent terms, providing a comparison of definitions, as condensed by Dorigato [11].

Tab. 1 – List of terms for plastics recycling and recovery (adapted from Dorigato [11]).

ASTM D5033	Equivalent ISO 15270	Other equivalent
primary recycling	mechanical recycling	closed-loop recycling
secondary recycling	mechanical recycling	downgrading
tertiary recycling	chemical recycling	feedstock recycling

According to Zheng and Suh [12], reducing the annual growth rate of plastic demand from 4% to 2% would result in a significant decrease in greenhouse gas emissions. Specifically, this reduction would reach 56% using the current energy mix and could reach up to 81% if low-carbon energy sources were utilized, compared to projections for the year 2050.

Among various plastics, polypropylene (PP) stands out as a widely used commodity polymer. Its success is attributed to its favorable price/performance ratio, excellent mechanical properties, and desirable optical characteristics. The extensive use of this polymer makes its recycling strategically crucial for the industry's environmental policy [13, 14].

Recycled plastic is more energy-efficient than virgin plastic [15] because, according to The Association of Plastic Recyclers [16], plastic recycling processes generally require less energy than the synthesis of plastic resins from basic chemicals. Specifically, the energy consumed (MJ/kg) to recycle PP is 3.0 times lower compared to its virgin counterparts. According to Zheng et al. [17], the use of recycled plastics instead of virgin plastics is advantageous both economically and environmentally, particularly for high-demand materials such as PP.

According to Li, Zhao, and Wang [18], the injection moulding (IM) technique represents a fundamental pillar in the manufacturing of plastic components, standing out for its automation, material efficiency, and ability to produce parts with complex geometries.

In IM, the properties of recycled materials can differ significantly from those of raw materials, making it more challenging to maintain the quality of moulded products [19]. The degradation of mechanical properties in products made from recycled plastic is a significant drawback that limits their use [20].

Based on the hypothesis that primary recycling and IM of PP cause its progressive degradation, mainly through chain scission, affecting its molecular structure, mechanical properties, and crystallization behavior, which in turn impacts the final product. This work of review investigates the degrading effects of primary recycling of PP during the IM process and their impact on the final product. Some authors such as Dananjaya et al. [21] and Ladhari et al. [22] reviewed previous works on the characterization of plastic products after multiple recycling cycles, however, this review seeks to be more specific regarding the raw material and its origin, with PP coming from primary recycling. Furthermore, this review study also considers types of screw elements.

2. Methodology

In order to fill the gap and validate or reject the research hyppostesis, a metodology called Currency, Relevance, Authority, Accuracy, and Purpose (CRAAP) was followed. This test helps to evaluate credibility and use value of sources, particularly when deciding whether to include them in a paper [23].

The papers avaliated and used during the review were obtained from Web of Science and Scopus databases. Additionally, the Backward Snowballing method was employed, which involved analyzing the reference lists of database articles to identify earlier works pertinent to the research [24].

3. Results

The search on the databases resulted in 9 of 3,493 and 2 of 20 for Web of Science and Scopus respectively. The Backward Snowballing approach made possible to recover more 9 papers. resulting in 20 papers of interest.

Table 2 presents a summary of the 20 recovered articles references (Ref.), listing their properties analyzed in the investigation of the phenomenon of interest. These properties encompass various types of analysis conducted by the authors, including thermal methods such as Thermogravimetric Analysis (TGA), Derivative Thermogravimetry (DTG), Differential Scanning Calorimetry (DSC), and Dynamic Mechanical Thermal Analysis (DMTA). Spectroscopic techniques include Fourier Transform Infrared Spectroscopy (FTIR), while rheological assessments involve Melt Flow Index (MFI) and Dynamic Rheological Testing (DRT). Mechanical evaluations feature Tensile (TT) and Impact tests (IT), Nanoindentation (NI), Creep testing (CT), and Microindentation hardness testing.(MHT), Optical examinations include Polarized Optical Microscopy (POM), Birefringence (BR), Wide-Angle X-ray Scattering (WAXD), and Scanning Electron Microscopy (SEM). Additionally, chromatography studies incorporate Gel Permeation Chromatography (GPC).

Furthermore, comprehensive overview of the conditions under which cyclic reprocessing was conducted, and the essential findings of the analysis—is systematically outlined in**Table 2**.

Tab. 2 – A review of existing studies on PP waste reprocessing from primary recycling.

Ref.	Analysis	Reprocessing condition	Key findings
[25]	MFI; GPC; FTIR	Extrusion was carried out in air at 260 °C using a 25 mm single-screw extruder (L/D = 20) at 100 rpm. The initial run processed powder, followed by pellets for up to 7 cycles.	PP degradation occurred through chain scission, decreasing average molecular weight (MW) from 418,400 to 249,300 in the first extrusion pass. Since the virgin polymer was extruded as a powder, surface oxygen absorption accelerated thermo-oxidative breakdown.
[26]	FTIR; GPC; MFI	A 2 mm single-screw extruder (L/D 24, 50 rpm) was used. The extrudate was quenched (5–10 °C), dried, pelletized, and reprocessed for up to 19 cycles at 240–270 °C.	No thermo-oxidative degradation occurred, with minimal carbonyl formation. Higher temperatures or repeated extrusion narrowed MW distribution. Degradation followed three regimes: none below 230 °C, temperature-dependent between 230–250 °C, and stable above 250 °C. Chain scission in high MW chains was the primary mechanism, without recombination, affecting extrusion parameters.
[27]	TT; DRT; GPC	PP was extruded with a temperature profile of 210 °C, 240 °C and 240 °C with a 0.35 g/s flow rate. After 7 cycles, it was ground using a blade mill.	Viscosity decreased after reprocessing due to reduced and narrower MW distribution. Mechanical analysis showed lower elongation at break with more recycling cycles.
[28]	FTIR; MFI; DSC; TT; IT	Samples underwent up to 10 reprocessing cycles, and then were injection-moulded at 200 °C, cooled for 20 seconds, and granulated. Injection time was 0.43 seconds, with a screw rotation rate of 200 rpm.	No chemical structure changes were observed, but increased MFI indicates chain scission as the primary degradation mechanism. During reprocessing, PP stays liquid or amorphous at lower temperatures, accelerating crystallization. Recycled PP has higher modulus, yield stress, and crystallinity but lower MW, elongation, and fracture toughness.
[29]	IT; SEM	Mechanical test specimens were recycled six times via IM and granulation per ASTM standards: 200°C melt, 20 s cooling, 594 bar injection pressure, 0.43 s injection time, and 200 rpm screw speed.	Recycling decreases spherulite size as more heterogeneous nuclei form than are destroyed by IM. Fracture analysis identifies crazing as the main mechanism in both virgin and recycled PP, with virgin PP absorbing more energy due to lower crystallinity and higher MW, improving toughness.
[30]	FTIR; GPC	Extrusion used a twin-screw extruder with five screw designs, a 5 kg/h feed rate, and 100 rpm screw speed. The material was reprocessed up to five times at 210–235 °C.	Repeated extrusions with aggressive screw profiles reduce PP's MW and polydispersity, increase carbonyl and unsaturation by scissioning long chains, and boost low MW fractions. A five-lobe, 90° kneading screw intensifies degradation, whereas left-handed conveyors limit oxidation by reducing oxygen contact.
[31]	TT; IT; MHT; SEM	PP was injection-moulded into tensile and notched bend specimens, and then granulated in a knife mill for six recycling steps.	PP's higher crystallinity gives it greater micro-hardness and a larger crack-tip plastic zone. SEM reveals that the skin layer, with lower crystallinity and affected by triaxial stress, displays reduced micro-hardness and a smaller plastic zone than the core.
[32]	MFI	PP was injection-moulded with 50 rpm screw and temperature of 265 °C. After, the samples were milled up to 4 times.	PP underwent progressive thermo-mechanical degradation, reducing MW with each processing cycle.
[33]	MFI; TT	PP was extruded at 230 °C into filaments, quenched in a 15 °C water bath, dried at 100 °C for 24 hours, and recycled four times.	MFI increased from 2.17 to 5.13 g/10 min after four extrusions, evidencing chain scission. Tensile strength dropped to 58% of the virgin sample, with a 20% decrease after the first extrusion.
[34]	MFI	PP was extruded (30 mm, L/D 32, 50 rpm), quenched (5–10 °C), dried, pelletized, and reprocessed for 5, 8, 12, 16, and 19 cycles at 240 and 270 °C.	Polymer degradation lowers viscosity and elasticity. Influencing factors include residence time distribution (via velocity profiles), stock temperature variation, deformation rate distribution, and degradation agents.
[35]	TT; IT; DSC	PP was extruded on a 30 mm single-screw extruder (L/D 32, 50 rpm), quenched (5–10 °C), dried, pelletized, and reprocessed 5, 12, and 19 cycles at 240–270 °C before IM.	High shear induces PP degradation via chain scission, reducing MW. After 19 cycles at 270 °C, crystallinity and heat of fusion rise, small-strain properties remain stable, and break properties sharply decline.

Tab. 2 – Continued.

Ref.	Analysis	Reprocessing condition	Key findings
[36]	DSC;TGA; FTIR, MFI; TT; SEM	PP was extruded, quenched, air-dried, and pelletized. Pellets were dried for 60 min, reprocessed up to six times at 200 °C, and injection-moulded into ASTM D638 type I samples at 190–220 °C using a 90-ton machine.	Thermo-mechanical degradation causes PP chain scission without oxidation, increasing MFI, crystallinity, and glass transition temperature while lowering melting, crystallization, and relaxation temperatures. Young's modulus and yield stress peak early then decline, and elongation at break continuously falls.
[37]	DSC; MFI; TT	Samples were reprocessed up to 15 times using a twin-screw extruder (150 rpm, 3 kg/h) at 205 °C. Thin sheets (15 mm × 2 mm) were extruded, cooled between calender rolls, and measured, with a 70 s extrusion residence time.	Reprocessing raises MFI and lowers zero-shear viscosity via chain scission, reducing MW while preserving the elastic modulus (~1200 MPa) and impact strength. After one cycle, the crystallization peak narrows, with further cycles yielding earlier crystallization (higher crystallization temperature) and increased crystallinity from smaller chains.
[38]	TGA; DTG; MFI	Samples were reprocessed 1, 3, and 5 times, dry mixed, fused at 180 °C (80 rpm), cooled to room temperature, and ground into small particles.	Thermo-degradation via chain scission reduced MW and increased MFI, with no oxidation detected. Thermogravimetric analysis showed a progressive decrease in activation energy over reprocessing cycles.
[39]	MFI; TT; POM	PP was extruded (34 mm twin-screw, 200 rpm) and reprocessed up to 5 cycles at 200 °C, then injection-moulded at 200 °C and 650 bar into ASTM Type V specimens, with five specimens tested per cycle (0, 1, 3, 5).	Viscosity decreased and MFI increased, indicating chain scission. TGA confirmed thermo-mechanical degradation producing smaller molecules, and tensile testing showed a slight decline in properties from extrusion cycles.
[14]	MFI; DSC; TT	Samples were dried at 80 °C for 4 hours, reprocessed up to five times, and injection-moulded at 210 °C (mould: 60 °C, 170 bar, 100 rpm, 5 bar back pressure). Two types were made for fluidity and tensile tests, with varied packing pressure switching points, pressures, times, and cooling durations.	Recycling raises MFI (stabilizing after four re-injections) and boosts crystallinity: chain breakage yields sharper peaks and smaller, uniform crystals that improve tensile strength by lowering the mould cavity peak and viscosity index. However, high screw speeds and melt temperatures may cause cracking, a risk that higher back pressure can mitigate.
[40]	FTIR, MFI, DSC, GPC	PP was extruded using a co-rotating twin-screw extruder at 400 rpm for five cycles at 240, 260, and 280 °C, with a 3-minute residence time per cycle.	PP degradation increases with higher temperatures and processing times, marked by a higher carbonyl index and MFI, and lower molecular weight, with critical effects at 240–260 °C and accelerated damage at 280 °C.
[41]	FTIR; DSC; DMTA; CT; POM; NI	Samples were dried at 90 °C for 4 hours, reprocessed up to 20 times, and extruded with a co-rotating twin-screw extruder at 90 rpm and 205 °C.	FTIR detected a 1735 cm^{-1} band (thermo-oxidative degradation). DMTA, NI and CT showed reduced stiffness and MW. DSC confirmed chain scission as the main mechanism. Recycling increased crystallization temperature by 9.1 °C (from 112.9 °C) and enhanced crystallinity, per POM.
[42]	TT; BR; WAXD	PP moulded into a 200×40×1 mm plate at 220 °C, split into far and near gate (NG). NG melt-spun at 230 °C through a 1 mm spinneret at 5.0 g/min, wound 350 cm below at 0.5–5.0 km/min without quench airflow.	At 1.0 km/min, recycled PP shows a downstream solidification shift from suppressed flow-induced crystallization due to reduced entanglement. At 5 km/min, its properties match virgin PP.
[43]	MFI; GPC	Two PP types PP_TF (thermoforming grade) and PP_MO (moulding grade) were IM and milled into flakes six times.	After six cycles, chain scission cuts PP molar mass by 39% (PP_TF) and 47% (PP_MO), causing an exponential MFI increase and liquid-like flow.

4. Conclusions

The hypothesis was supported and the objective of the study was fulfilled. The review highlights the complex degradation mechanisms of PP during multiple reprocessing cycles. Chain scission is identified as the predominant degradation, leading to a progressive reduction in molecular weight and an increase in the MFI. Despite thermo-mechanical stress, no significant signs of oxidation were observed, suggesting that thermo-oxidative degradation has little impact on PP's deterioration.

Recycling changes PP's physical and mechanical properties. It increases crystallinity, stiffness (modulus), and strength (yield stress) while decreasing flexibility (elongation), toughness, and viscosity. As the polymer is exposed to heat repeatedly, its structure evolves, leading to faster crystallization and changes in how crystals form. The temperature at which crystallization begins shifts, and the way spherulites (structures that develop as the polymer solidifies) are formed also changes. These transformations demonstrate how recycling influences the material's ability to crystallize and solidify.

Mechanical degradation, rather than chemical alteration, explains PP's declining elasticity, viscosity, and micro-hardness, particularly in recycled samples subjected to high shear forces. While virgin PP demonstrates superior strength and toughness, recycled variants behave in a consistent way during degradation, following a well-documented pattern. The narrowing molecular weight distribution confirms that degradation preferentially affects high-molecular-weight chains. The impact of screw profile aggressiveness further elucidates the role of processing parameters in controlling material deterioration.

4.1 Future work

Ultimately, recycling preserves some PP properties, but cumulative chain scission reduces break stress and fracture resistance. Future work should optimize processing (with stabilizers and advanced techniques) to mitigate degradation and enhance sustainability.

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