SPECIAL SYMPOSIA | CIRCULAR ECONOMY OF POLYMERS

Unlocking the Potential of Recycled Polypropylene in Food Packaging

N. Krempl, M. Fruehwirth, Z. Shahroodi, C. Holzer,
E. Pinter, E. Jahn, V.H. Gabriel and F. Aschermayer

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

December 2024

Unlocking the Potential of Recycled Polypropylene in Food Packaging

N. Krempl, M. Fruehwirth, Z. Shahroodi, C. Holzer, E. Pinter, E. Jahn, V.H. Gabriel and F. Aschermayer[1]

Abstract: This study aims to enhance recycled polypropylene (rPP) for yogurt cups, promoting sustainable resource use and waste reduction. Analysis of sorting, recycling, and decontamination shows mechanical recycling of PP effectively generates new products. Experiments reveal pre-sorted, hot-washed, and color-sorted rPP has superior quality and is processable up to 100% recyclate content. Adding a masterbatch additive improves oxidative stability and reduces degradation. Biological, chemical, and sensory analyses confirm rPP cups match virgin PP in odor and appearance, with no mutagenic substances detected. This demonstrates the feasibility of high-quality rPP in food packaging, supporting sustainable practices in the circular economy.

Keywords: Mechanical Recycling, Polypropylene, Sustainable Packaging, Circular Economy, Material Characterization, Industrial Testing, Quality Assurance, Research Collaboration

1 The authors N. Krempl, M. Fruehwirth, Z. Shahrood and C. Holzer are affiliated with the Institute of Polymer Processing at the University of Leoben in Austria. E. Pinter is affiliated with the Department of Packaging, Recycling and Dangerous Goods, at the Austrian Research Institute for Chemistry and Technology in Austria. The authors E. Jahn and V.H. Gabriel are affiliated with the Packaging and Resource Management at the University of Applied Life Sciences in Austria. Finally, F. Aschermayer is affiliated with Greiner Packaging International GmbH in Austria.

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

Introduction

Guided by the Circular Economy Action Plan, the European Union aims to achieve climate neutrality by 2050. This includes making all plastic packaging recyclable and recycling 55% of plastic packaging and 60% of municipal waste. In 2022, 54 million tons of plastics were processed in Europe, with packaging leading at 21.1 million tons. Of the total processed polymers, 32 million tons were collected and nearly 38% of packaging waste was recycled [1].

Several European countries, including Austria and Germany, have banned landfill plastics, leading to higher recycling rates despite increased incineration. Polypropylene (PP) is the most commonly used plastic, constituting about 19% of the total. Austria, for example, has a PET beverage bottle recycling rate of about 73% and is expected to reach 90% by 2030 with its new deposit system.

Austria's yellow bag system for collecting plastic packaging aims to improve sustainability. Collected waste is being sorted, but challenges persist. For example, the trade sector's choice between recycling or using virgin packaging significantly impacts market dynamics. Advanced sorting technologies are needed to handle complex materials and outdated regulations hinder progress.

The Pack2theLoop project, funded by the Austrian Research Promotion Agency (FFG), involves 40 companies represented by ecoplus, the Business Agency of Lower Austria and four research teams collaborating to develop quality-assured recyclates from post-consumer packaging for new packaging applications. This article focuses on rPP for food packaging, ensuring the recyclates meet stringent quality requirements. The project, conducted on an industrial scale, aims to improve recycling processes and material quality, enhancing the sustainability and efficiency of the packaging sector.

Materials and Methods

In order to improve the recycling processes and material quality, the first step involved establishing a status quo by examining a specific Austrian material stream using state-of-the-art sorting and recycling methods and technologies in 2021 (Material Loop 1). Building on these initial insights, the project progressed to a second phase that focused on enhanced quality improvement steps (Material Loop 2).

Material Loop 1

Initial industrial sorting employed the Austrian output fraction ARA 414 (a mix of PP and PS), whose input and output were analyzed in detail in manual sorting trials. The sorted materials underwent industrial recycling with water separation, comparing cold vs. hot washing methods. At the Starlinger facility, decontamination involved extrusion, filtration, degassing, and hot air treatment.

Biological, chemical, and sensory analyses evaluated quality and contamination levels for potential conversion into yogurt cups.

Sorting trials in separate collection were conducted manually at three locations from 2021 to 2024 to assess the potential of PP recycling from Austrian household plastic packaging waste. Detailed analysis focused on PP lightweight packaging collected from yellow bags or bins. Initially, waste was separated by material type, followed by sorting based on criteria including food/ non-food, shape (cup, hollow body, etc.), color, labeling(L) (direct printing, sleeve, L>50%, L<50%, etc.) and closure (with/without). Quantities were documented by weight and number of units to evaluate recycling feasibility.

Initial findings showed significant inhomogeneity in colored rPP recycling stream, promoting characterization of hot-washed, color-sorted rPP granules decontaminated to PET standards.

Material Loop 2

Given its availability, PP with the material specification 324-0 polypropylene [2] from Der Grüne Punkt, Germany, was selected for Material Loop 2. In contrast to Material Loop 1, this phase included additional pre-sorting of white and natural-colored rigid PP for food contact to enhance input purity. The recycling process comprised float-sink separation using saline solution and cold water, followed by drying, dedusting, flake sorting, and a hot wash for thorough cleaning. Decontamination at the Starlinger facility involved extrusion, filtration, degassing, and hot air treatment of the granules.

Proof of concept through an industry study

The study analyzed the processability of decontaminated material from Loop 2 using an inline thermoforming process involving extrusion and thermoforming of monolayer films. The process utilized standard settings designed for virgin cups, with the material demonstrating excellent processability and requiring minimal adjustments. Despite a high melt flow index (MFI) of 17 g/10 min (230°C, 2.16 kg), for the decontaminated rPP, up to 100% rPP content did not present any thermoforming issues. However, challenges in long-term serial production could potentially arise.

Biological, chemical, and sensory analyses

The analyses included migration studies using HS-SPME-GC/MS for volatile substances assessment. Toxicological evaluations were conducted via the miniaturized Ames test to detect DNA-reactive mutagens in decontaminated recyclate and yogurt cup samples. Samples underwent migration in 95% ethanol at 60°C for 10 days, followed by analysis or concentration for methodological purposes. Additionally, sensory evaluation by a trained panel assessed odor profiles to compare with virgin samples.

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

Material quality improvement taking into account multiple processing

Here, a twin screw extruder and a combination masterbatch (MAXITHEN® PP7AB4750AO) from Gabriel Chemie GmbH were conducted to enhance the property profile of decontaminated recyclate within the circular economy framework. The extrusion process employed a Leistritz Extrusion GmbH ZSE 27 MAXX compounder operating at 350 rpm, with processing parameters set at 210°C and L/D ratio of 44.3 wt.-% of the masterbatch additive was incorporated during the initial cycle. Pelletization was performed using an Econ GmbH underwater pelletizer (type EUP 50 D). The resulting compounds underwent ten extrusion cycles and was subsequently analyzed thermally, rheologically, and by gel permeation chromatography to assess the material properties.

Thermal analyses involved measuring the oxidation induction time (OIT) using differential scanning calorimetry (DSC 1, Mettler Toledo GmbH, Switzerland). Granules were uniformly pressed into 1 mm plates for consistent sample surfaces. Samples with a diameter of 5 mm and weighing 10 mg were prepared and heated under inert gas (N_2) at 185°C for 2 minutes, followed by exposure to oxygen for 60 minutes to induce oxidation.

Rheological measurements (MCR702, Anton Paar GmbH, Austria) were conducted in a nitrogen atmosphere, evaluating complex viscosity, storage, and loss moduli across an angular frequency range of 0.1–500 rad/s at 185°C and 1% deformation. The crossover point of storage and loss moduli was utilized to qualitatively assess average molecular weight and molecular weight distribution, indicating material degradation [3].

Gel permeation chromatography (GPC) was employed to quantitatively analyze the results obtained from rheological measurements. The analysis utilized 1,2,4-trichlorobenzene (TCB) as the solvent, which was stabilized with butylated hydroxytoluene (BHT) and n-heptane. High-temperature GPC coupled with an IR detector was conducted at 160°C, employing three mixed bed columns and a guard column with a flow rate of 1 mL/min. The analysis, calibrated using narrow PP standards, spanned 40 minutes per sample to determine molecular weight distribution. Prior to analysis, samples were prepared by mixing 8 mg of each with a flow marker and TCB, followed by dissolution, filtration, and measurement using an external filtration system.

Results and Discussion

Sorting trials in separate collection

Manual sorting resulted in approximately 8 wt.-% rigid PP packaging in Austrian separate waste collection of household lightweight packaging waste. A significant share of approximately three quarters represents food packaging in the PP sorting. PP food cups account for 1.4 wt.-% in the yellow bag, which are mainly white coloured with 71 wt.% Figure 1. In addition, approximately 12 wt.-% of the identified PP food cups have an aluminium platine attached.

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

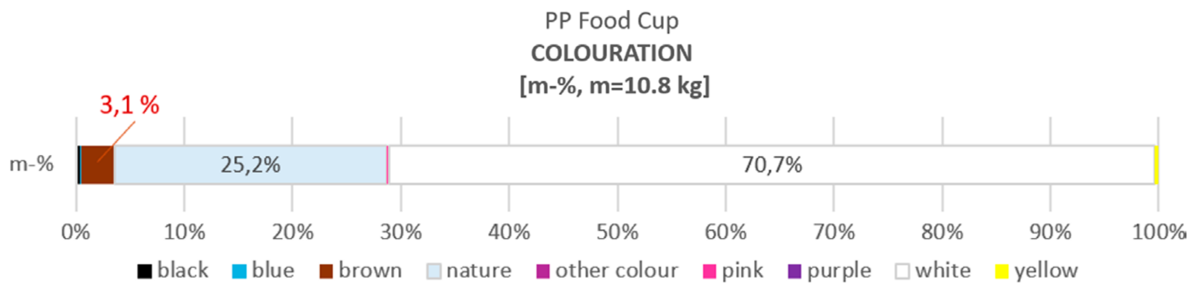

Figure 1. Packaging waste analysis of PP food cups concerning coloration of the main packaging body. 10.8 kg were identified as PP food cups in the sorting input of which 71 wt.-% were white and a quarter transparent.

Industrial Study

In the industrial evaluation of decontaminated rPP production, several key insights into its properties and performance were identified. A high melt flow index of 17 g/10 min (30°C, 2,16 k) was found also to be suitable for up to 100% rPP content, although ongoing production may require continuous monitoring to address potential long-term issues. Material processability was excellent, with minimal adjustments needed, though rPP cups exceeding 30% rPP sometimes exceeded weight specifications. Introducing 30% rPP into virgin material showed acceptable variations in color and odor, indicating minor sensory differences were manageable at this concentration. Uniform lower diameter limits were hardly achieved, but the deviations remained small and acceptable. In general, large fluctuations were found in the measurement protocol of the cup dimensions. The material's top load capacity remained unaffected up to 70% rPP, indicating robustness and macroscopic impurities (Figure 2) probably require better filtration during decontamination.

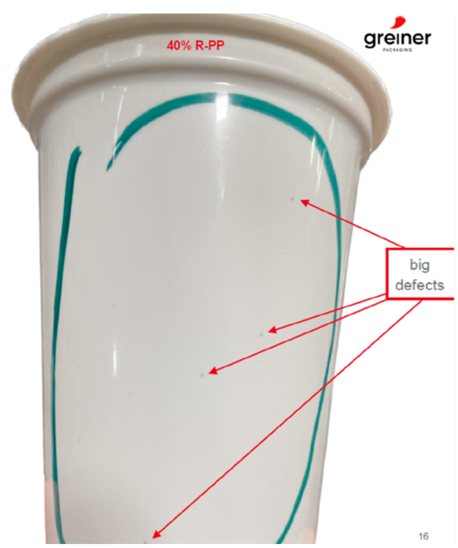

Figure 2. PP yoghurt cup with 40 w.% rPP and 60% vPP with several marked black spots [4].

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

Biological, chemical, and sensory analyses confirmed the potential of decontaminated granulates for use in food packaging. Under worst case migration conditions, chemical screening revealed no deviations compared to virgin material, and bioassay results indicated no DNA-reactive mutagenic effects. The odor of the decontaminated PP resembled that of virgin PP granulates, supporting its application in cup production. Further analyses of 100% rPP cups, considered most critical, showed no adverse effects in bioassays and no anomalies in chemical composition across varying recycling contents (30%, 60%, 100% rPP). Although fewer high volatile substances were detected compared to virgin PP, attributed to effective decontamination processes, sensory analysis indicated no perceptible differences with increasing recyclate content, affirming comparable odor profiles to cups made from virgin PP.

Material quality improvement taking into account multiple processing of the decontaminated recyclate by adding additive involved systematic exploration within the circular economy framework. The material underwent rigorous testing to explore its capabilities through multiple processing cycles, focusing on degradation behavior. Analysis centered on the impact of a masterbatch, integrating oxidation stabilization and compatibilizer, on OIT and molecular mass, illuminating the material's degradation process.

Addition of 3 wt% of the masterbatch extended OIT by a factor of 4.5, maintaining 2.5 times higher stability than unstabilized samples even after 10 cycles (Table 1).

Table 1. OIT at a temperature of 185°C for rPP and rPP with 3 wt% masterbatch.

Cycle	$OIT_{185°C}$ in min		factor x
	rPP	rPP+3wt.% MB	
1x comp	9	42	4,5
5x comp	8	23	3,0
10x com	7	17	2,5

The rheological findings were consistent with those from GPC. Figure 4 illustrates the correlation between these methods for pure rPP and the rPP masterbatch compound. Over successive cycles, all compounds showed gradual material degradation, as evidenced by the upward and rightward shift of the crossover points (Gc) of G' and G'' (left diagram of Figure 3). The polydispersity index (PDI) in the right diagram of Figure 3, which indicates the breadth of molecular weight distribution, revealed a 28% degradation in the rPP compound after 10 cycles due to molecular chain breakdown. Incorporating 3 wt% masterbatch reduced this degradation by approximately 5%.

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

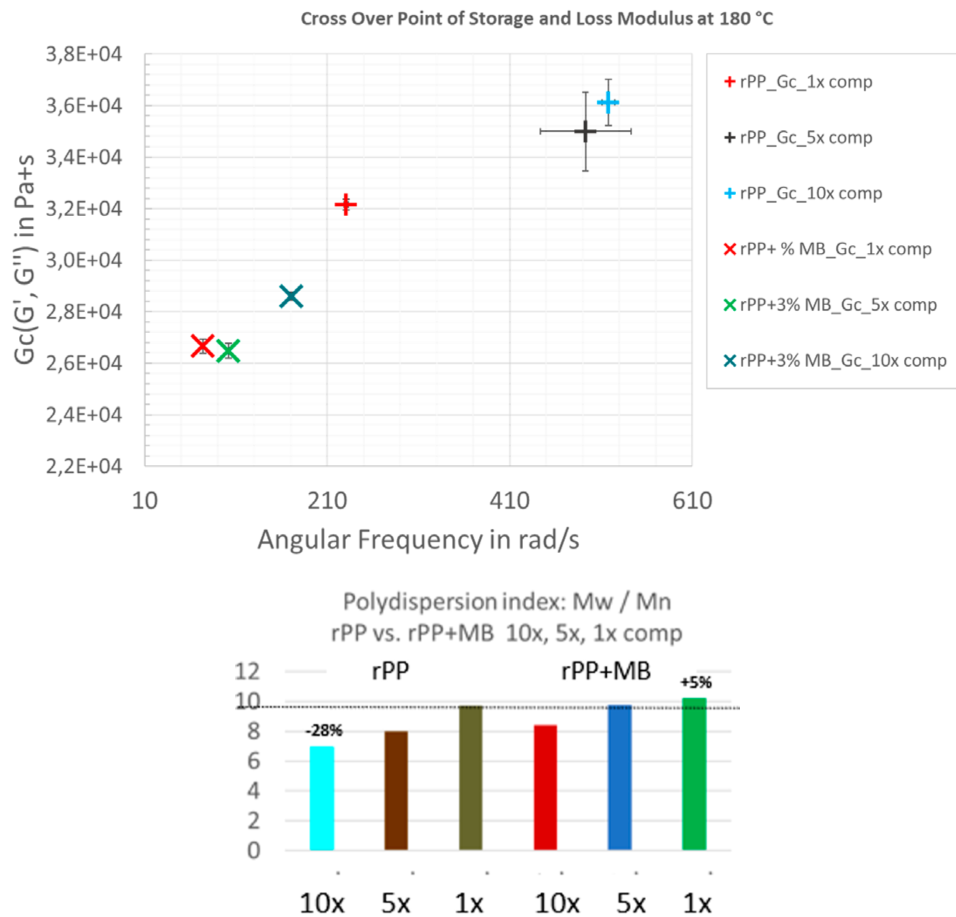

Figure 3. Above: Crossover Point Gc (G', G'') as a function of angular frequency ω at a temperature of 185°C. Below: Polydispersity for rPP without and with masterbatch (MB).

Conclusion

This study conclusively supports the use of rPP in food packaging, aligning with the European Union's Circular Economy Action Plan goals. Through advanced sorting, recycling, and decontamination methods, rPP demonstrated suitability for stringent quality standards without compromising processability, even at high content levels (up to 100%). Sensory evaluations indicated minimal odor differences compared to virgin PP cups, supporting consumer acceptance. Biological and chemical analyses confirmed the safety of decontaminated rPP, showing no mutagenic effects or chemical deviations under worst case migration scenarios. The study highlights the effectiveness of additives like masterbatch in enhancing oxidative stability and mitigating material degradation during multiple processing cycles, crucial for scaling up rPP adoption in packaging. Continued innovation and regulatory support will be vital to further advance sustainable practices and achieve higher recycling rates across Europe.

DOI: https://doi.org/10.51573/Andes.PPS39.SS.CEP.2

Acknowledgments

We gratefully acknowledge the funding support from the Austrian Research Promotion Agency (FFG) for the Pack2theLoop project. We also extend our appreciation to the 40 companies represented by ecoplus, the Business Agency of Lower Austria, and the four research teams whose collaboration was instrumental in the success of this project. Their contributions have significantly advanced our understanding and capabilities in developing quality-assured recyclates from post-consumer packaging for new packaging applications.

References

1. N.N., Plastics Europe, Conversion Market & Strategy GmbH, and nova-Institut, *The Circular Economy for Plastics: A European Analysis*, 2024. [Online]. Available: https://plasticseurope.org/knowledge-hub/the-circular-economy-for-plastics-a-european-analysis-2024/. Accessed: Jun. 20, 2024.
2. N. Wallus, Der Grüne Punkt, "Raw material fraction specification 324-0 Polypropylene," Version 1.00.0001, Doc. No. DOC-23-50686, 2024. [Online]. Available: https://www.gruener-punkt.de/fileadmin/Dateien/Downloads/PDFs/Rohstofffraktionsspezifikationen2024/April/DOC-23-50686_-Rohstofffraktionsspezifikation_324-0_Polypropylen-_v1.00.0001.pdf.
3. N.N., Thermo Fisher Scientific – Materials & Structural Analysis, "Optimizing Polymeric Materials with Rheological Analysis," 2023. [Online]. Available: https://www.azom.com/article.aspx?ArticleID=20979. Accessed: Aug. 28, 2023.
4. F. Aschermayer, Greiner Packaging International GmbH, "PP yoghurt cup with 40 w.% rPP and 60 % vPP with several marked black spots," *Pack2theLoop User Meeting Presentation*, Nov. 29, 2023.