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WHAT YOU ARE, TAKES YOU FAR

Doctoral Dissertation
Doctoral Program in Materials Science and Technology (38th Cycle)

From waste to resources: Tailored recycling strategies for polyolefins

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Summary

Plastic itself is not the problem; rather, the way plastic waste is managed represents the real challenge. Polymers are indispensable materials in modern society, yet their sustainability is critically undermined by ineffective end-of-life management and the limited performance of recycled materials. This thesis addresses these challenges by focusing on the mechanical recycling of polyolefins - specifically polypropylene (PP) and high-density polyethylene (HDPE) - which dominate plastic production. The work is grounded in a critical analysis of current plastic production, waste management, and recycling practices, highlighting how, despite increasing regulatory pressure and ambitious policy targets, the plastics economy remains largely linear. It is now well established that meaningful sustainability improvements must rely on enhanced circularity rather than indiscriminate material substitution. Within this context, mechanical recycling is identified as the most mature and energy-efficient recycling route.

The experimental work systematically investigates the thermo-mechanical degradation of polypropylene during repeated extrusion cycles, revealing that PP degradation is dominated by random chain scission, leading to progressive reductions in molecular weight, without significant oxidative modification of the polymer. To counteract these effects, a reactive repair additive is evaluated under conditions representative of both pre-consumer and post-consumer mechanical recycling. The results demonstrate that the additive is effective in mitigating viscosity loss, partially rebuilding molecular weight, and inducing controlled modifications of the macromolecular architecture.

Beyond short-term processing behavior, the long-term performance of recycled polypropylene is assessed through photo-oxidative aging experiments. The evolution of chemical structure, thermal response, and mechanical properties is systematically analyzed, providing clear evidence that the introduction of the repair additive is also able to maintain adequate mechanical properties for long periods under photo-oxidative degradation. In this way, the useful life of films produced with recycled plastic can be extended, greatly reducing the loss of elongation at break and stress at break even when exposed to solar UV light and condensation cycles.

This work further explores the mechanical recycling of high-density polyethylene, whose degradation behavior is characterized by the coexistence of chain scission, branching, and crosslinking reactions, leading to highly heterogeneous microstructures. By tailoring processing conditions, these degradation-induced reactions are deliberately exploited to promote long-chain branching. As a result, recycled HDPE with enhanced melt strength and increased ductility is obtained, enabling its use in processing technologies dominated by elongational flow.

In addition, the thesis investigates the functional upgrading of recycled PP through the incorporation of different flame-retardant systems. For each formulation, rheological behavior, morphology, and fire retardancy are systematically compared with those of virgin PP-based systems. The results demonstrate that effective flame retardancy can be achieved in recycled matrices, while also revealing how polymer degradation influences the efficiency and stability of flame-retardant additives.

Finally, the end-of-life mechanical recycling of flame-retarded PP is examined by subjecting an intumescent flame-retardant system to multiple extrusion cycles. The evolution of rheological, mechanical, morphological, and fire-retardant properties during repeated reprocessing highlights the combined effects of polymer degradation and additive redistribution. These findings provide critical insights into the recyclability of functionalized plastics and underline the importance of designing flame-retardant systems that remain compatible with mechanical recycling across multiple life cycles.

Overall, unlike conventional re-stabilization approaches, which merely slow down further degradation, the strategies developed in this PhD thesis actively enable structural recovery and functional upgrading. The results reported in this PhD dissertation prove that through targeted microstructure control and functional design, recycled polyolefins can achieve enhanced performance, extended application potential, and higher added value. These findings demonstrate that mechanical recycling can evolve from a downcycling practice into a robust technological pillar for a circular plastics economy.