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Simulation of a Real Recycling Scenario for an HDPE-Based Packaging

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ABSTRACT

Several factors still limit the achievement of effective mechanical recycling strategies for polyolefins. In particular, the thermo-mechanical degradation undergone during reprocessing, the different degradation forms experienced during the service life, as well as the cross-contaminations resulting from non-fully accurate sorting technologies, cause the obtainment of recyclates with heterogeneous microstructure, which significantly affect their final properties, often limiting their possible future applications. This study evaluates the combined effects of aging and cross-contamination for high-density polyethylene (HDPE) containing low amounts of polypropylene (PP) and polyethylene terephthalate (PET) as contaminants. HDPE systems were subjected to photo-oxidative or thermo-oxidative treatments and reprocessing to simulate real recycling scenarios. Results show that PP and PET contamination significantly reduce HDPE ductility under thermo-oxidative conditions, while exerting minimal impact on photo-oxidized materials.

1 | Introduction

The transition to a circular plastics economy is increasingly recognized as a solution to mitigate the environmental impacts of plastic waste. By incorporating repair, reuse, and recycling strategies, plastics can be reintroduced into production cycles, reducing reliance on landfilling and incineration, which contribute to soil pollution, marine litter, and greenhouse gas emissions [1]. Currently, mechanical recycling dominates plastic waste processing. However, challenges persist in maintaining the material's economic value during recycling. Degradation processes such as photo-oxidation and thermo-mechanical stress alter polymers' microstructure, affecting their molecular weight and processability, as well as their mechanical properties. All these issues often result in downcycling processes, involving the re-utilization of recycled polymers for applications with lower value added than their virgin counterparts [2].

Besides, due to ineffectiveness of the sorting techniques exploited for the selection of polymeric materials prior to mechanical recycling, recycled materials often contain mis-sorted polymers, whose presence further reduces the quality of recyclates. This issue is particularly relevant for polyolefins, such as HDPE and PP, characterized by similar density. In fact, it has been shown that recycled HDPE typically contains 2%–13% PP contamination [3]. Although a few studies have examined the impact of polyolefin contamination on the final properties of recyclates, the combined effects of such cross-contamination and the degradation experienced by polymers during their service life and reprocessing remain largely unexplored.

This work aims to investigate the interplay between aging and cross-contamination on HDPE mechanical recycling. Simulating a typical recycling process for an HDPE-based bottle, HDPE systems containing small amounts of PP and PET contaminants were subjected to aging treatments and reprocessed. The interactions between cross-contamination, aging treatments, and the thermo-mechanical degradation experienced by the material during reprocessing were analyzed, focusing on the functional and microstructural changes induced in HDPE, as well as on the mechanical behavior of the investigated systems.

2 | Results and Discussion

The results of the spectroscopic characterization highlighted that the presence of PP as a contaminant in HDPE lowers carbonyl index (C.I.) values (calculated from the ATR-FTIR spectra as the ratio between the intensity of the carbonyl peak at 1720 cm^{-1} and that of a reference peak) compared to pristine HDPE, both before and after photoaging, but has minimal impact under thermo-oxidative conditions. According to the literature, the stabilization effect of PP can be attributed to its faster radical formation during degradation as compared to HDPE [4]. As a result, PP acts as a sort of sacrificial oxidizable phase, exerting a stabilizing effect toward the oxidation of HDPE. Under thermo-oxidative conditions, this effect is less pronounced. Otherwise, the contemporary presence of PP and PET leads to higher C.I. values with respect to HDPE. However, it should be considered that the absorption peaks in the carbonyl range exploited for monitoring the degradation of HDPE are already present in the ATR-FTIR spectrum of pristine PET. Therefore, also in this case, it is possible to assess that the presence of cross-contaminants minimizes the degradation of HDPE. The microstructural variation of the investigated systems subjected to aging and reprocessing was monitored through rheological analyses. The obtained results, coupled with the morphological observations through SEM, demonstrated that the introduction of PP and PET leads to the obtainment of immiscible blends, whose final microstructure strictly depends on the effect of the specific aging treatment on the viscosity of the HDPE matrix phase. In particular, the sample HDPE/PP/PET_ph showed significant morphological alterations, induced by both the photo-oxidation and the reprocessing, resulting on the achievement of a more refined morphology as compared to its non-aged counterpart. Conversely, the thermo-oxidized samples exhibited a quite unmodified morphology as compared to their non-aged counterparts, and only some alterations of the main size and shape of the dispersed phase inclusions or of their extent of dispersion within the matrix were noticed.

The chemical and microstructural changes in HDPE caused by aging and reprocessing in the presence of cross-contamination can significantly affect its mechanical performance. The performed tensile tests (whose main results are reported in Table 1) revealed that while elastic modulus and tensile strength remain largely unaffected, ductility is dramatically reduced. Before aging, the addition of PP as a contaminant has minimal impact on tensile properties. However, the simultaneous presence of PP and PET significantly decreases elongation at break, indicating that PET causes notable embrittlement of the HDPE matrix.

As a consequence of the photo-oxidative treatment and reprocessing, HDPE-based samples exhibit a dramatic embrittlement, which is even more severe for cross-contaminated materials. This behavior can be associated with the heterogeneous microstructure of the aged and reprocessed materials induced by the different phenomena (namely, chain scission, crosslinking, and branching) undergone by HDPE during the photo-oxidative treatment and the reprocessing. On the other hand, the thermo-oxidative treatment has a negligible effect on the tensile properties of pristine HDPE. A very different behavior is observed for the samples containing cross-contamination. In this case, in fact, similarly to photoaged samples, the thermo-oxidative treatment and the reprocessing induced a decrease in the elongation at break as compared to HDPE without PP and PET contaminants.

3 | Conclusions

In this work, the interplay between the degradation experienced by HDPE during its service life and reprocessing, and the presence of PP and PET cross-contamination were assessed, aiming at simulating the conditions undergone by the polymer during a mechanical recycling process. Starting from a typical HDPE bottle composition, materials with cross-contaminants were subjected to photo-oxidative or thermo-oxidative aging, followed by reprocessing. The functional and microstructural modifications induced by the aging treatments and reprocessing were assessed and discussed, also considering their impact on the mechanical properties of HDPE. The obtained results revealed that cross-contaminants, particularly in photo-oxidized samples, reduced the formation of oxygen-containing functional groups, indicating a mitigating effect on the functional degradation. The tensile tests demonstrated that under thermo-oxidative conditions, PP and PET cross-contaminations have a detrimental role for the ductility of reprocessed HDPE, thereby potentially affecting any possible reutilization of the material for the same application as its virgin counterpart. At variance, in the case of the photo-oxidative treatment, the main aspect remarkably reducing the HDPE elongation at break is the severe degradation undergone by the polymer, while the presence of cross-contaminants exerts a marginal effect.

4 | Experimental Section

4.1 | Materials

HDPE ERACLENE BB82 (Versalis), PP Moplen HP500 N (LyondellBasell), and PET PPK FR (Plastipak Italia) were used.

4.2 | Processing

Three systems were produced: (i) pristine HDPE, (ii) HDPE with 10 wt.% PP, (iii) HDPE with 10 wt.% PP and 2 wt.% PET. The compounding was carried out using a Process 11 (Thermo Fisher) twin-screw extruder (screw rotation speed: 100 rpm, temperature profile: 230-230-230-240-240-240-250-250-250°C from hopper to die).

4.3 | Aging

(i) Photo-oxidative treatment by subjecting the specimens to UV irradiation in a SEPAP 12/24 (Atlas) apparatus for 300 h; (ii) thermo-oxidative treatment at 80°C in an oven at RH 100%

TABLE 1 | Mechanical properties of investigated HDPE-based systems.

	Elastic modulus (MPa)	Tensile strength (MPa)	Elongation at break (%)
HDPE	1279.1 ± 13	25.8 ± 0.4	936.3 ± 25.5
HDPE/PP	1282 ± 47	25.5 ± 0.5	908.9 ± 30.9
HDPE/PP/PET	1274 ± 27	24.3 ± 1.2	20.8 ± 4.3
HDPE_ph	1150 ± 27	22.8 ± 0.5	84.5 ± 38.6
HDPE/PP_ph	1116 ± 70	22.9 ± 0.6	31.4 ± 8.2
HDPE/PP/PET_ph	1212 ± 11	24.6 ± 0.4	12.7 ± 1.5
HDPE_th	1220 ± 148	26.2 ± 0.4	834.1 ± 28.1
HDPE/PP_th	1256 ± 39	23.6 ± 1	268.9 ± 91.3
HDPE/PP/PET_th	1311 ± 102	25.7 ± 0.4	34.9 ± 8.8

for 300 h. After the aging treatments, all the systems were reprocessed through the ThermoFischer Process 11 twin-screw extruder.

4.4 | Characterization

Frequency sweep tests were performed using an ARES (TA Instruments) rheometer under N_2 atmosphere at 230°C. Spectroscopic characterization was carried out through FTIR-ATR Frontier (Perkin Elmer) equipped with a diamond crystal. Scanning electron microscopy EVO 15 by Zeiss was employed for morphological characterization. Tensile tests were carried out using a 5966 Instron dynamometer equipped with a 2 kN cell load and 250 N clamps.

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Conflicts of Interest

The authors declare no conflicts of interest.

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