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Reactive Degradation and Upgrading of Recycled Polypropylene Using Nexamite Additives: Effects on Rheological, Thermal and Mechanical Properties

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Abstract

To reduce the environmental impact associated with fossil-based materials, mechanical recycling represents a key strategy, although it inevitably leads to a decline in rheological, thermal, and mechanical properties due to polymer chain degradation. This study investigated the effect of controlled degradation and subsequent reprocessing of polypropylene (PP) using reactive additives from the Nexamite family. Degradation was induced by adding Nexamite R202 at concentrations of 0.5%, 1%, and 2%, while reprocessing was carried out using Nexamite R203 combined with an antioxidant package. The results show that peroxide addition promotes progressive chain scission, leading to reduced thermal stability and mechanical performance. A moderate peroxide content (1%) provided the best balance between improved processability and mechanical performance, showing high ductility and impact toughness. Subsequent reprocessing with Nexamite R203 enabled a significant recovery of the degraded material's properties. Formulation PP REC 8.2 exhibited increased melt viscosity, slight thermal stabilization, and a marked improvement in elongation at break while maintaining stress values comparable to the starting degraded material. Overall, the results demonstrate that a strategy based on controlled degradation followed by targeted reprocessing can effectively tune processability and partially restore the performance of recycled polypropylene, offering promising opportunities for the upgrading of recycled PP for higher-value applications.

Keywords: plastic recycling; sustainable polymers; reactive extrusion; mechanical performance

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1. Introduction

The production of plastic materials has increased significantly in recent decades [1,2], showing the highest growth among all bulk materials [3] and accounting for 4.5% of global greenhouse gas emissions [4]. The intensive production and consumption of plastics has progressively raised awareness of environmental issues related to polymer waste management and fossil resource consumption [5,6]. In this context, reducing environmental impacts can be achieved through the use of recycled polymeric materials [7], aiming to lower CO_2 and greenhouse gas emissions [8] while promoting a circular economy model. Life cycle assessment (LCA) has shown that the production of recycled polypropylene reduces CO_2 emissions by 22.6% compared to the corresponding virgin material [9]. Ref. [10] highlighted that mechanical recycling of polymers reduces environmental impacts by

97% compared to traditional virgin material production. Moreover, mechanical recycling has a lower environmental footprint compared to chemical recycling and is also more cost-effective [11,12]. Among the most industrially relevant polymers, polypropylene (PP) stands out due to its good mechanical properties [13,14], high versatility [15,16] and ease of processing via major industrial techniques such as extrusion and injection molding [17,18]. However, mechanical recycling of polypropylene [19] still faces several challenges [20,21], particularly in high-performance applications. One major limitation is the degradation of the material's mechanical properties, resulting in lower quality final products [22]. During processing and reprocessing, the material undergoes repeated thermal, mechanical, and oxidative stresses that trigger polymer chain degradation [23,24]. Degradation leads to chain scission, molecular weight reduction, and worsening of rheological and mechanical properties [25]. Consequently, recycled material exhibits inferior properties compared to its virgin counterpart [26]. Chain scission significantly affects rheological behavior, as reduced molecular weight increases the melt flow index (MFI) [27–30]. The increase in MFI corresponds to a reduction in melt viscosity, as confirmed by [27,31]. Several studies have attempted to mitigate degradation effects. Ref. [32] reported that the addition of titanium dioxide (TiO_2) to polypropylene reduces degradation and improves thermal stability, but simultaneously decreases melt viscosity, thus limiting optimal results [33] studied polypropylene degradation and found that adding virgin polymer can help counteract degradation, but does not improve stability if the material is already degraded. Phenolic antioxidant packages effectively inhibit degradation while present, but once consumed, the material degrades rapidly. Ref. [34] analyzed unstabilized polypropylene and material stabilized once at the beginning with phosphite and phenolic additives; despite initial benefits, after five cycles 50% of the additives had already degraded, showing this is not an optimal solution. Ref. [35] studied the improvement of recycled material properties using additives and stabilizers, finding that adding virgin polymer or antioxidants alone does not significantly improve performance. In contrast, chain extenders produce excellent results for polyesters like PET and polyamides (PA), as degradation generates reactive groups (OH, COOH, NH_2) that can chemically react. For polyolefins such as polypropylene (PP) and polyethylene (PE), these reactive groups are absent, limiting chain repair. Therefore, currently there is no fully effective solution to improve mechanical, thermal, and chemical properties of polyolefins, particularly polypropylene. A possible approach is the use of a specific reactive additive during extrusion, Nexamite R202, designed to induce controlled degradation of polypropylene (PP), with the dual purpose of reducing melt viscosity and improving material flow during processing, while simultaneously promoting chain scission and modifying the molecular architecture of the polymer. In this way, an intermediate degradation state can be achieved, characterized by increased chain mobility while preserving sufficient structural integrity. This controlled degradation step is particularly relevant within the proposed two-step strategy, as it prepares the material for the subsequent reactive reprocessing stage. As reported in [36], a similar Nexamite additive (R201) enables controlled degradation, generating a new polymer architecture rather than regenerating virgin material. Another key aspect is the possibility of reprocessing degraded material, which represents a significant technological challenge. By using specific additives such as Nexamite R203 in combination with suitable antioxidant packages, the negative effects of degradation can be mitigated, restoring a balance between viscosity, thermal stability, and mechanical performance. These additives rebalance the molecular structure and limit further oxidation during processing. As a result, a partial recovery of the material properties can be achieved. The present work aims to systematically analyze the effect of controlled degradation of polypropylene by adding Nexamite R202 at different concentrations (0.5%, 1%, and 2%), evaluating its impact on the rheological,

thermal, mechanical, and chemical properties of the degraded material. In this context, the study introduces the concept of a controlled degradation window, identifying an optimal degradation level that maximizes the effectiveness of subsequent reactive reprocessing. Subsequently, a reprocessing phase of the previously degraded polypropylene is investigated using Nexamite R203 in combination with an antioxidant package, with the aim of restoring and tuning material performance for industrial applications. The objective of this two-step strategy is not simply to reverse degradation, but to create an optimized balance between processability and final material performance. Controlled degradation improves melt flow and chain mobility, while subsequent reactive reprocessing allows partial recovery of mechanical integrity and thermal stability. This combined approach may provide advantages that are difficult to achieve through a single treatment step. The developed formulations are characterized using a wide range of experimental techniques, including melt flow index (MFI) determination, differential scanning calorimetry (DSC), capillary rheometry, FTIR spectroscopy, tensile and impact testing, and heat deflection temperature (HDT) measurements. This approach enables the correlation between additive-induced structural modifications and macroscopic material behavior, providing a practical framework for optimizing polypropylene recycling and reprocessing strategies.

2. Materials and Methods

2.1. Material

The polypropylene grade employed in this study was C715-12NHP, purchased from Braskem (São Paulo, Brazil), with a Melt Flow Rate (230 °C/2.16 kg) of 12 g/10 min. To induce material degradation, Nexamite R202 (Nexam Chemical Holding AB, Lomma, Sweden) was added. This is a reactive masterbatch designed to regulate the flow behavior of polypropylene, both recycled and virgin, by modifying its viscosity. Table 1 shows the formulations investigated in this study.

Table 1. Formulations with Braskem polypropylene and Nexamite R202.

Component	PP REC 7	PP REC 8	PP REC 9
Polypropylene Braskem C715-12NHP (%)	99.5	99	98
Nexamite R202 (%)	0.5	1	2

In a second step, the degraded material was reprocessed with Nexamite R203 (Nexam Chemical Holding AB, Lomma, Sweden) and an antioxidant package (ADK STAB A-611, ADEKA Corporation, Tokyo, Japan), with the formulations reported in Table 2.

Table 2. Formulation with aged polypropylene and reprocessing additives.

Component	PP REC 8.1	PP REC 8.2	PP REC 8.3
PP REC 8	99.5	95.8	95.76
Nexamite R202 (%)	0	0	0.04
Nexamite R203 (%)	0	4	4
ADK STAB A-611 (%)	0.5	0.2	0.2

The concentration of Nexamite R203 (4 wt.%) was selected within the recommended range (2–5 wt.%) reported in the technical datasheet, as a representative intermediate value to ensure effective reactive rebuilding while maintaining processability. The exact chemical composition of Nexamite R202 and R203 is proprietary and not fully disclosed by the manufacturer; therefore, the discussion of their effect is based on their functional behavior as reported in the literature and supported by the experimental results. The experimental

procedure consisted of two main steps. First, virgin polypropylene was dry blended with different amounts of Nexamite R202 and processed by twin-screw extrusion to induce controlled degradation, producing the PP REC 7, PP REC 8, and PP REC 9 formulations. R202 was therefore added before the first extrusion step to promote controlled chain scission and modify the rheological behavior of polypropylene. Based on the first-stage results, PP REC 8 was selected for reactive reprocessing, as it showed the best balance between melt flow and mechanical performance. In the second step, PP REC 8 was dry blended with Nexamite R203 and the antioxidant package, then reprocessed by twin-screw extrusion. R203 was added only during this second stage. For PP REC 8.3, a small amount of R202 was also added during reprocessing together with R203 and the antioxidant package. The overall experimental workflow adopted in this study, including the controlled degradation step and the subsequent reactive reprocessing stage, is schematically illustrated in Figure 1.

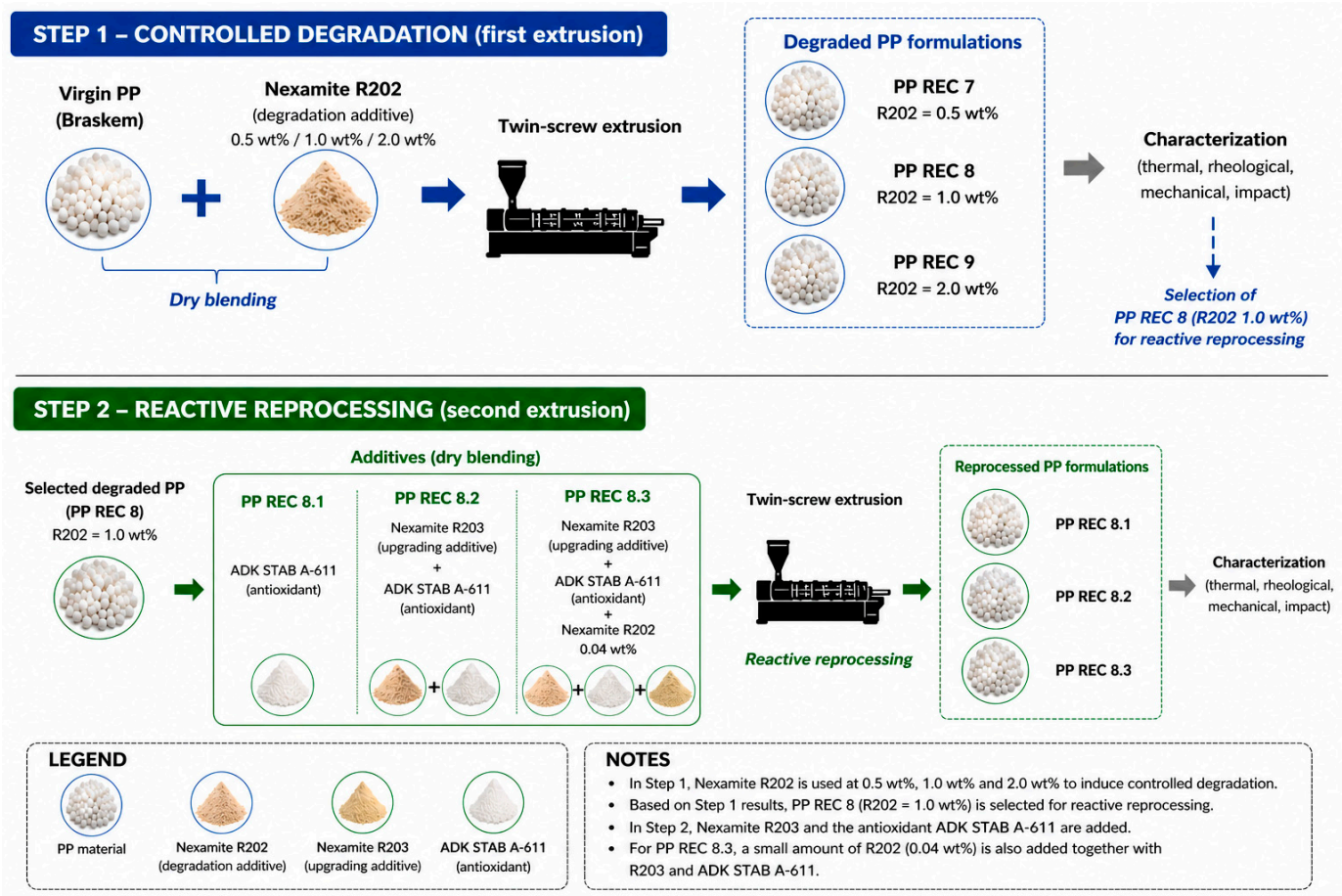


Figure 1. Schematic representation of the two-step experimental procedure adopted.

2.2. Twin Screw Extrusion

Compounding was carried out using the Leistritz ZSE 27 iMAXX co-rotating twin-screw extruder (Leistritz Extrusionstechnik GmbH, Nürnberg, Germany) with a screw diameter of 27 mm and an L/D ratio of 48, which allows for homogeneous mixing of the materials. The raw materials, previously premixed manually, were fed into the first section of the extruder via a gravimetric feeder. The extrusion system was equipped with ten independently controlled heating zones (T1–T9 and die head). Since the formulations showed different rheological behaviors depending on the degradation and reprocessing stage, specific thermal profiles were adopted for each formulation in order to ensure stable melt flow and regular strand formation. The barrel temperature profiles used for PP REC 7, PP REC 8, PP REC 9, and for the reprocessed formulations PP REC 8.1, PP REC 8.2, and

PP REC 8.3 are reported in Tables 3–6, respectively. The same processing conditions were adopted for PP REC 8.1, PP REC 8.2, and PP REC 8.3. The selected temperature settings allowed proper melting, mixing efficiency, and dimensional stability of the extruded strands, while minimizing further thermo-mechanical degradation during processing.

Table 3. Thermal profile PP REC 7 formulation of the extruder.

Machine's Parameters										
Thermal Profile	T1	T2	T3	T4	T5	T6	T7	T8	T9	Head
	180	182	185	188	190	190	188	185	182	180

Table 4. Thermal profile PP REC 8 formulation of the extruder.

Machine's Parameters										
Thermal Profile	T1	T2	T3	T4	T5	T6	T7	T8	T9	Head
	170	170	175	185	190	190	188	180	175	172

Table 5. Thermal profile PP REC 9 formulation of the extruder.

Machine's Parameters										
Thermal Profile	T1	T2	T3	T4	T5	T6	T7	T8	T9	Head
	160	170	175	175	180	185	180	178	170	165

Table 6. Thermal profile PP REC 8.1, 8.2 and 8.3 formulation of the extruder.

Machine's Parameters										
Thermal Profile	T1	T2	T3	T4	T5	T6	T7	T8	T9	Head
	155	160	165	170	170	180	180	175	170	155

The adopted thermal profiles were based on a humpback configuration, characterized by an initial increase in temperature along the extrusion barrel followed by a progressive decrease toward the die. In the first section of the barrel, the temperature was gradually raised to promote efficient feeding, melting, and homogenization of the polymer. In the second section, the temperature was reduced toward the extrusion head in order to increase melt strength and elasticity at the die exit, thus improving strand stability and pelletizing performance. Depending on the formulation and its rheological behavior, slight adjustments of the temperature settings were applied, as reported in Tables 3–6. This approach ensured stable processing conditions while limiting excessive thermo-mechanical degradation of the recycled material. Figure 2 shows the extruder die with the polymer strand exiting (a), the strand entering the cutting system (b), and the final pellets (c).

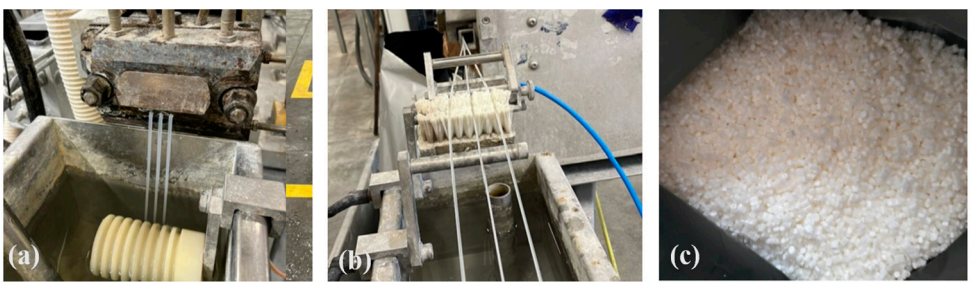


Figure 2. (**a**) Polymer strand exiting the die, (**b**) strand entering the pelletizing system, (**c**) final pellets.

The screw speed used during processing was 280 rpm, with a melt pressure of 52 bar. The melt temperature at the die exit was 152 °C, lower than the profile temperature, confirming the decreasing thermal profile toward the head and allowing the cooling of the strand.

2.3. Pellet Characterization

2.3.1. Melt Flow Index (MFI)

The Melt Flow Index (MFI) test is commonly used to estimate melt flow behavior and indirectly assess viscosity changes. In this case, a load of 2.16 kg is applied, and the test is performed at 230 °C. The test was carried out using the Melt Flow Indexer Model 1221 from AMSE (Turin, Italy) according to ISO 1133 [37]. For each formulation, three independent samples were tested, and the reported values correspond to the average of the measurements.

2.3.2. Differential Scanning Calorimetry (DSC)

Among the characterization techniques employed for pellet characterization, differential scanning calorimetry (DSC) was used to evaluate thermal properties and crystallinity. This test allows the determination of the polymer's thermal properties and degree of crystallinity. Analyses were conducted using a DSC3 (Mettler Toledo, Columbus, OH, USA). All samples were placed in aluminum crucibles for the measurements, with sample weights ranging between 8 and 12 mg. Nitrogen was used as the purge gas at a flow rate of 50 mL/min, in accordance with ISO 11357 [38]. The following formula, shown in Equation (1), was used to calculate crystallinity:

$$x_{CL} = \frac{\Delta H_{mi} - \Delta H_{ci}}{\rho_i * \Delta H^0_{mi}} \quad (1)$$

where ΔH_{mi} and ΔH_{ci} are the melting and cold crystallization enthalpies during the heating scan, respectively, while ΔH^0_{mi} represents the enthalpy of fusion for a 100% crystalline material. ρ_i is the fraction of the polymer for which the crystallinity is to be measured. Since no cold crystallization peak was observed for the analyzed polypropylene samples, ΔH_{ci} was considered equal to zero in this study, and the crystallinity was calculated using only the melting enthalpy contribution.

The degree of crystallinity was calculated using a reference melting enthalpy of 207 J/g for fully crystalline polypropylene, as commonly reported for isotactic PP in DSC analyses [39].

2.3.3. Rheological Analysis

The rheological properties of the blends were analyzed using a CEAST SmartRHEO R20 capillary rheometer (Instron, Pianezza, Turin, Italy). The die used had a diameter of 1 mm and a length-to-diameter ratio of 20. Measurements were performed at 180, 190, and 200 °C. For each test, a preheating period of 300 s with a compacting force of 50 N was applied. The tests were conducted at 180, 190, and 200 °C with shear rates ranging from 50 to 2000 s^{-1}.

2.3.4. Fourier Transform Infrared Spectroscopy (FTIR)

The infrared absorption spectra were recorded using a Jasco FT/IR-6600 spectrometer (JASCO Europe s.r.l., Cremella, Lecco, Italy) in ATR (attenuated total reflectance) mode. Measurements were performed in the range of 600–4000 cm^{-1}, with a resolution of 0.4 cm^{-1} and 128 scans. The analysis was carried out on pellets for all formulations. For each formulation, three independent samples were analyzed, and the reported carbonyl index

values correspond to the average of the measurements. FTIR analysis was used to assess the degree of polypropylene degradation through the carbonyl index, as reported in Equation (2) [40]:

$$Carbonyl\ Index = \frac{Area\ under\ band\ 1800 - 1650\ cm^{-1}}{Area\ under\ band\ 1500 - 1420\ cm^{-1}} \quad (2)$$

The index is defined as the ratio between the intensity of the carbonyl band (~1715 cm^{-1}) and a stable reference band of the polymer [41].

2.4. Sample Characterization

2.4.1. Tensile and Izod Test

Mechanical characterization was performed through tensile testing according to ISO 527 [42] using a Shimadzu AGS-X dynamometer (Shimadzu Corporation, Kyoto, Japan), a universal testing machine equipped with a 5 kN load cell and a maximum guaranteed error of 1%. Tests were conducted on dog-bone specimens at a speed of 10 mm/min, in accordance with ISO 527. Impact testing was carried out using an Izod XJUD pendulum from AMSE (Turin, Italy), with a 5 J hammer for the tests. Specimens were produced by injection molding using a RI.PI machine according to ISO 527 standards.

2.4.2. Heat Deflection Temperature Test

The test to determine the heat deflection temperature (HDT), that is, the temperature at which the polymer under a specified load—in this case 0.45 MPa—reaches a standard deformation. The test was carried out according to ASTM D648 [43] using an HDT/Vicat tester manufactured by AMSE (Turin, Italy).

3. Results and Discussion

3.1. MFI Test

Table 7 reports the Melt Flow Index (MFI) values of polypropylene formulations containing Nexamite R202 at concentrations of 0.5%, 1%, and 2%. Results showed that doubling the peroxide content increased the MFI by 19% in the case of PP REC 8, while PP REC 9 the increase reached 66%. The addition of peroxide caused significant polymer chain degradation, resulting in a decrease in molecular weight and, consequently, a marked reduction in melt viscosity, which accounts for the increased flow.

Table 7. MFI values for the PP REC 7, PP REC 8, and PP REC 9 formulations.

Formulations	PP REC 7	PP REC 8	PP REC 9
Melt Flow Rate (g/10 min)	32 ± 0.4	38 ± 0.5	53 ± 0.7
Nexamite R202 (%)	0.5	1	2

The increase in melt flow index observed with increasing Nexamite R202 content is consistent with the findings of [44] for post-consumer polypropylene, who reported that repeated thermo-mechanical reprocessing cycles induce polymer chain scission and molecular weight reduction, resulting in an increased melt flow rate. These results suggest that the MFI increase can be directly linked to the material's molecular degradation. Similarly, Ref. [28] highlighted that repeated mechanical recycling cycles cause degradation in polyolefinic materials, leading to an increase in melt flow index and a decrease in viscosity, indicating progressive chain scission that compromises processability and mechanical properties. The observed increase in MFI is indicative of molecular weight reduction due to chain scission.

3.2. DSC

Figure 3 shows the DSC curves for PP REC 7, 8, and 9, respectively. The melting temperature during the first heating shows limited variations among the formulations, with a slight decrease for PP REC 8 compared to PP REC 7 and PP REC 9. The crystallization peak also decreases with increasing percentage of Nexamite R202 in the formulation. This indicates a greater difficulty of the material to crystallize during cooling, with polymer chains that appear more degraded and less regular. During the second heating, the melting temperature remains stable for PP REC 7 compared to the first heating, while it shows a slight decrease for PP REC 8 and a larger decrease for PP REC 9, the latter due to the onset of thermal degradation of the material.

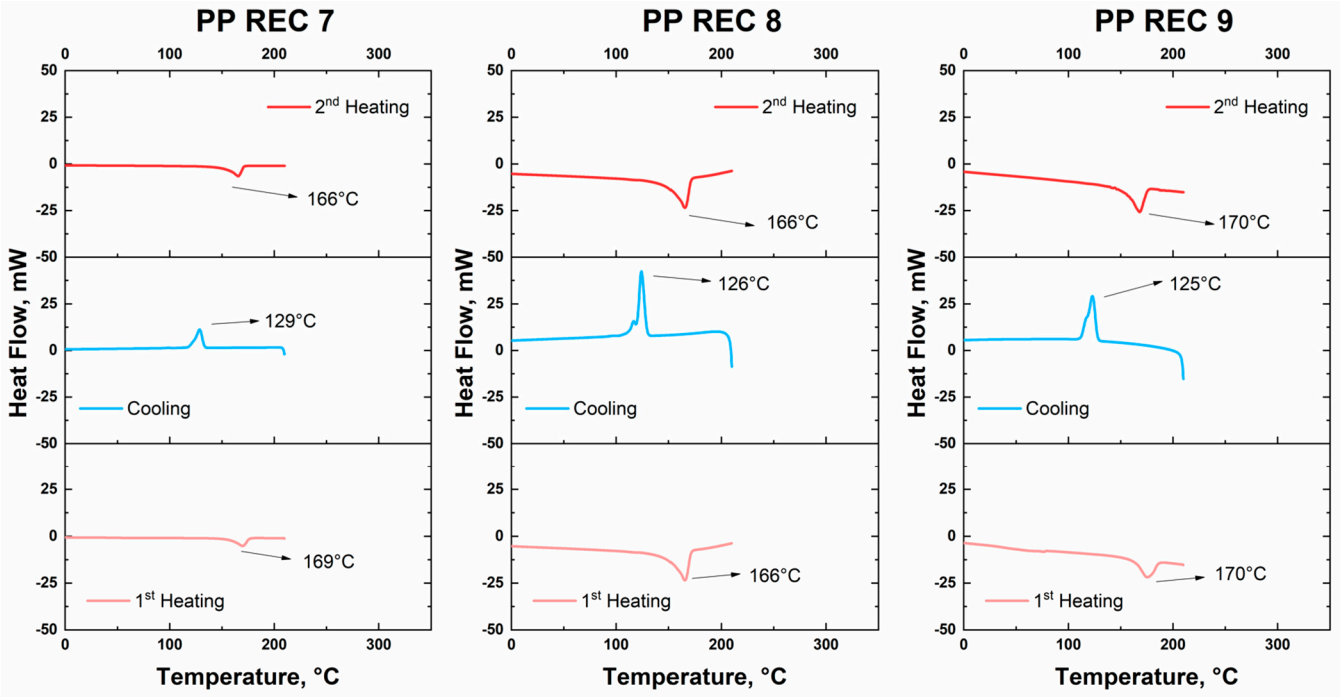


Figure 3. DSC curves for the PP REC 7, PP REC 8, and PP REC 9 formulations.

Table 8 reports the melting temperatures from the first and second heating, the crystallization temperature, and the degree of crystallinity. The latter shows a decrease, although moderate, indicating that the increase in peroxide content leads to a reduction in the crystallinity of the material.

Table 8. Degree of crystallinity, melting temperature, and crystallization temperature of the PP REC 7, PP REC 8, and PP REC 9 formulations.

	PP REC 7	PP REC 8	PP REC 9
Crystallinity degree	X_{CL} =0.26	X_{CL} = 0.25	X_{CL} = 0.24
Melting temperature (1st heating)	170 °C	166 °C	170 °C
Crystallization temperature	129 °C	126 °C	125 °C
Melting temperature (2nd heating)	166 °C	171 °C	170 °C

3.3. Capillary Rheometry

Figure 4 shows the shear viscosity curves for PP REC 7, 8, and 9, respectively. In all cases, the initial viscosity is rather high and decreases rapidly as the shear rate increases, showing a typical shear-thinning behavior. The curves obtained at the three test temperatures (180 °C, 190 °C, and 200 °C) present moderate differences, especially in the low

shear-rate region, highlighting a certain thermal sensitivity of the viscosity. Overall, the base recycled polypropylene maintains a regular rheological response, although it shows a temperature dependence in the initial stages of flow. The PP REC 9 formulation confirms the pseudoplastic behavior of the material: the viscosity is high at low shear rates and progressively decreases as the shear rate increases. The curves obtained at the three temperatures (180 °C, 190 °C, and 200 °C) show a more pronounced temperature dependence compared to PP REC 8, particularly in the low shear-rate regime. With increasing temperature, a significant reduction in viscosity is observed, indicating a strong thermal sensitivity of the material. This behavior may be attributed to possible molecular degradation or to an increase in fluidity typical of non-additivated recycled polypropylenes.

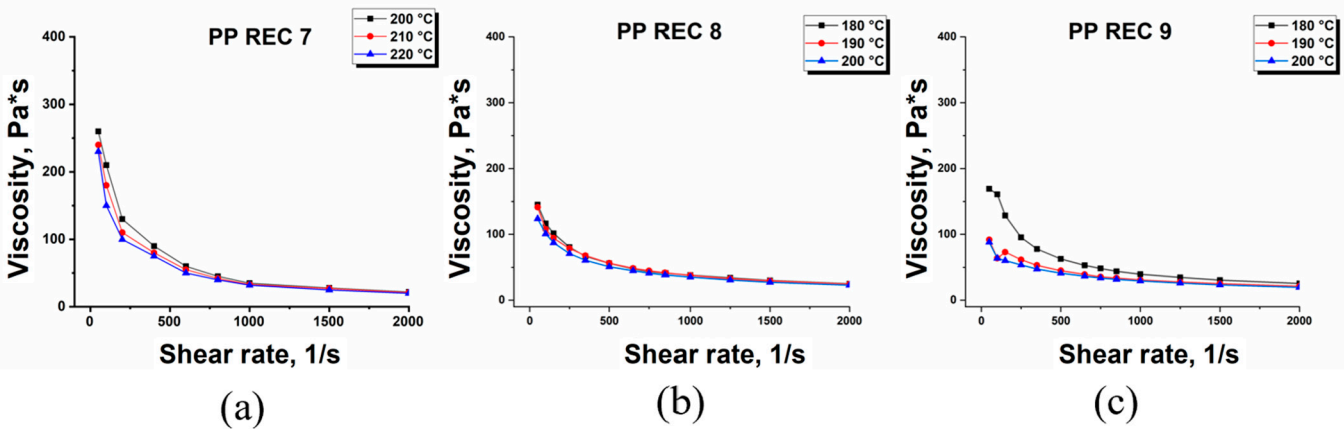


Figure 4. Shear viscosity curves of (**a**) PP REC 7, (**b**) PP REC 8, and (**c**) PP REC 9 formulations.

3.4. FTIR

Figure 5 shows the results related to the PP REC 7, PP REC 8, and PP REC 9 formulations. The profiles obtained are very similar, despite the addition of Nexamite R202. In fact, the characteristic bands of polypropylene appear in all the formulations:

- ~2950–2860 cm^{-1}: stretching bands C–H typical of aliphatic chains.
- ~1450 and 1375 cm^{-1}: bending of the CH_2/CH_3 group
- ~1160–975 cm^{-1}: C–C stretching and CH_3 rocking of the semicrystalline polymer.

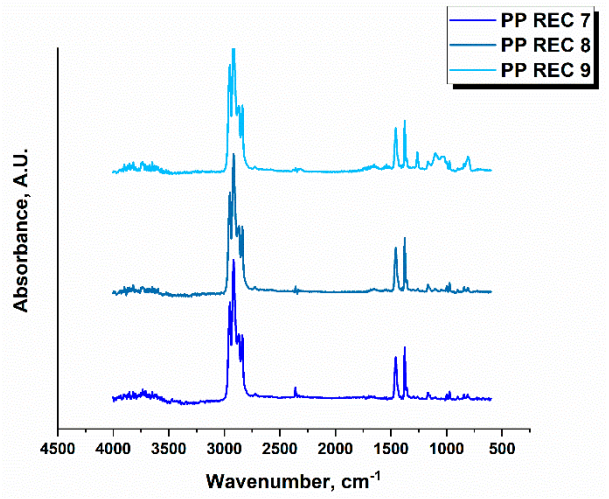


Figure 5. FTIR spectra of PP REC 7, PP REC 8, and PP REC 9 granules.

Table 9 shows the carbonyl index values for the different formulations. The recycled PP samples exhibit increasing carbonyl index values of 0.30 (PP REC 7), 0.45 (PP REC 8), and 0.86 (PP REC 9). This trend indicates a progressive increase in the level of oxidation,

consistent with greater chemical degradation of the material. In particular, CI values higher than 0.3 are generally associated with significantly oxidized polypropylene, in which chain scission mechanisms and the formation of oxygenated species become relevant [45].

Table 9. Carbonyl index for the formulations PP REC 7, PP REC 8 and PP REC 9.

Formulations	Carbonyl Index
PP REC 7	0.30
PP REC 8	0.45
PP REC 9	0.86

3.5. Tensile, Izod and HDT Test

Figure 6 shows the stress–strain curves for the formulations REC 7 (a), REC 8 (b), and REC 9 (c), respectively. The PP REC 7 formulation exhibits the highest average stress values of approximately 20.5 MPa. After yielding, the curve shows a pronounced strain-softening phenomenon, and fracture occurs at relatively low strains, roughly between 15 and 20%. The curves are not fully overlapping, reflecting some material heterogeneity that may be attributed to a non-uniform distribution of microstructural properties, or to varying levels of degradation or oxidation. The stress–strain diagram for PP REC 8 shows slightly lower average stress values (19.5 MPa) but higher strain values, reaching up to 60–65% for the most ductile sample. The smooth and continuous shape of the stress–strain curve suggests better structural integrity of the material, associated with lower degradation and more effective stabilization during the extrusion process. Consequently, PP REC 8 also exhibits the highest toughness, defined as the area under the curve, and therefore a greater capacity to absorb energy before fracture. Overall, the combination of good strength and high ductility makes PP REC 8 the formulation with the best overall mechanical performance. The PP REC 9 formulation shows the lowest stress and strain values, indicating that a high peroxide concentration (2%) induces more pronounced material degradation, resulting in significant reduction in mechanical properties. Additionally, the curve lacks a significant plateau, indicating poor plastic deformation capacity. This behavior is consistent with a more degraded material, in which polymer chain scission has reduced molecular weight and ductility, or with the presence of structural defects or oxidation and aging phenomena that promote premature fracture initiation. Table 10 reports the average tensile properties of the investigated formulations, expressed as mean ± standard deviation.

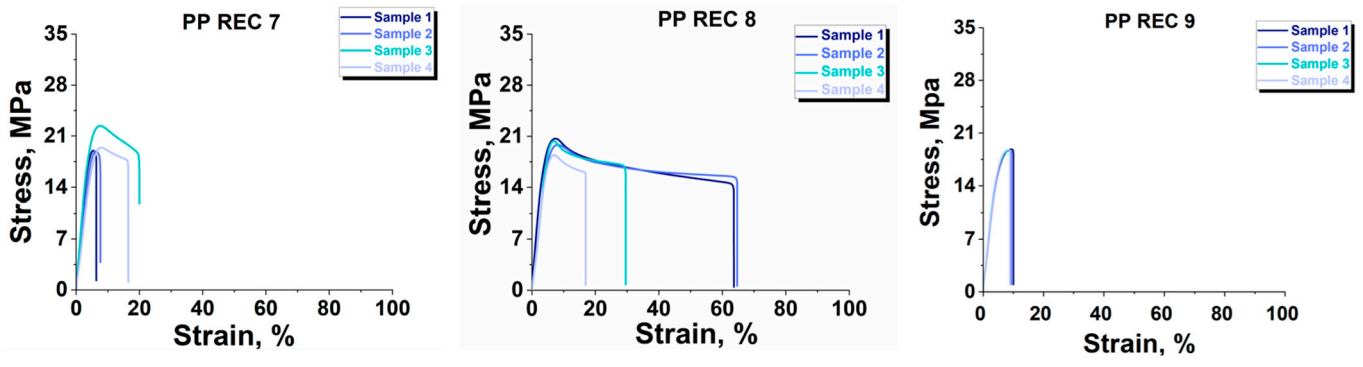


Figure 6. Stress–strain curves of (**a**) PP REC 7, (**b**) PP REC 8, and (**c**) PP REC 9 formulations.

Table 10. Strain and stress values for each analyzed formulation.

Mechanical Properties	PP REC 7	PP REC 8	PP REC 9
Strain (%)	16 ± 2.1	40 ± 8.5	10.5 ± 1.4
Stress (MPa)	20.5 ± 0.6	19.5 ± 0.7	17.5 ± 0.5

The reduction in tensile strength and elongation at break observed with increasing peroxide content is consistent with the findings of [46], who attribute the decline in mechanical properties of recycled polymers to polymer chain scission and the consequent reduction in molecular weight. Figure 7 shows the box plot of the impact energy distribution, and Table 11 reports the corresponding energy values for the PP REC 7, 8, and 9 formulations, allowing an immediate comparison of the median, variability, and presence of any outliers. The test was performed on four replicates. The formulation with 1% peroxide appears to perform best in terms of energy impact, with values higher than both PP REC 7 and PP REC 9, the latter showing poorer values due to material degradation. Regarding data variability, it is higher for PP REC 7 but lower for PP REC 9, indicating good data uniformity.

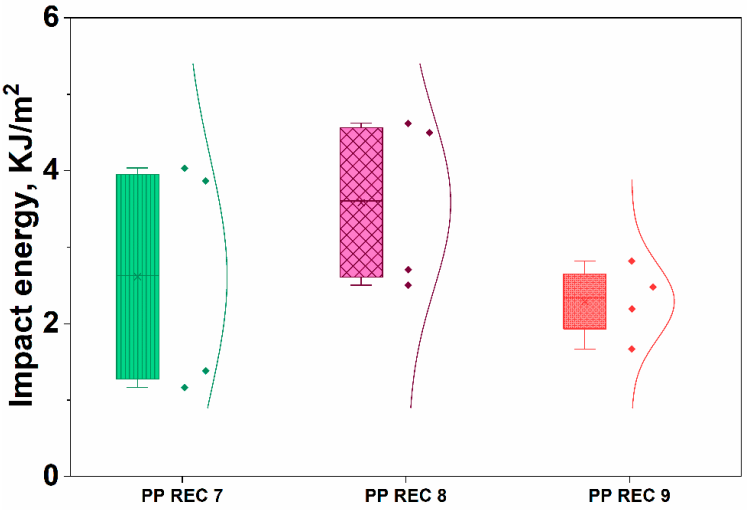


Figure 7. Box plot of the impact energy distribution.

Table 11. Impact energy values for the PP REC 7, PP REC 8, and PP REC 9 formulations.

Formulations	PP REC 7	PP REC 8	PP REC 9
Peroxide quantity (%)	0.5	1	2
Impact Energy (kJ/m^2)	2.6125 ± 1.45	3.5825 ± 1.08	2.291 ± 0.52

3.6. HDT Test

Table 12 shows the average softening temperatures of the material, based on three replicates. In this case, PP REC 7 exhibits the highest heat deflection temperature values compared to the formulations with 1% and 2% peroxide, respectively. In particular, the latter shows lower values due to material degradation.

Table 12. Heat deflection temperature values.

Formulations	Temperature (°C)
PP REC 7	82.9
PP REC 8	81.8
PP REC 9	80.7

3.7. Identification of the Controlled Degradation Window

The relationship between degradation level and mechanical performance reveals a clear non-linear trend, suggesting the existence of a controlled degradation window. As shown in Figure 8, the elongation at break increases with increasing degradation up to an intermediate level (PP REC 8), defining a controlled degradation window in which an optimal balance between processability and mechanical performance is achieved. Beyond this point, a significant decrease is observed at higher degradation levels (PP REC 9). Each data point represents the average of five specimens, ensuring good statistical reliability of the observed trend despite the limited number of degradation levels investigated. This behavior indicates that an optimal balance between processability and mechanical performance can be achieved only within a specific range of degradation. At low degradation levels (PP REC 7), the material retains a relatively high molecular weight, limiting chain mobility and resulting in reduced ductility. Conversely, excessive degradation (PP REC 9) leads to extensive chain scission and structural damage, causing a marked deterioration of mechanical properties. The intermediate condition (PP REC 8) represents an optimal state in which partial chain scission enhances molecular mobility without compromising the integrity of the polymer network, leading to improved elongation at break. This behavior can be attributed to the competition between chain scission mechanisms, which enhance molecular mobility, and excessive degradation, which reduces entanglement density and mechanical integrity. These results support the existence of a controlled degradation window, within which the material exhibits enhanced processability while maintaining sufficient structural integrity for effective mechanical performance. This concept is particularly relevant for optimizing recycling strategies, as it highlights the importance of carefully controlling the degradation level prior to any reprocessing step. These findings demonstrate that the effectiveness of subsequent reactive reprocessing is strongly dependent on the initial degradation state, making the identification of a controlled degradation window a key parameter for optimizing polypropylene recycling strategies.

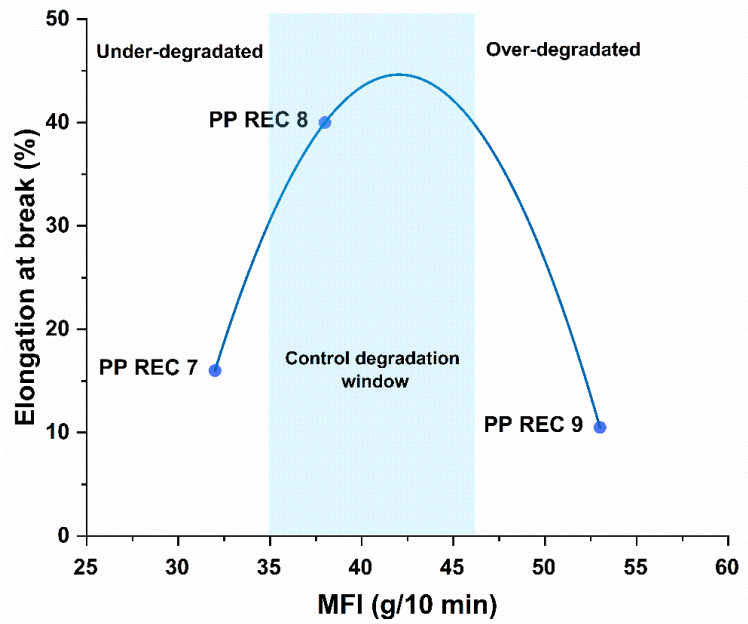


Figure 8. Relationship between degradation level (MFI) and elongation at break for PP REC 7, PP REC 8, and PP REC 9. The shaded region identifies the controlled degradation window, where an optimal balance between processability and mechanical performance is achieved.

4. Reactive Reprocessing of Degraded Polypropylene

4.1. MFI, DSC, FTIR Test and Rheology

Based on the identification of the controlled degradation window, the formulation PP REC 8 was selected for subsequent reactive reprocessing. Table 13 reports the MFI values of the formulations subjected to reactive reprocessing. In the first case, reprocessing with the antioxidant alone results in a decrease in the flow index by approximately 50%, indicating an increase in the material's viscosity. The addition of 4% Nexamite R203 allows the polymer to maintain higher viscosity values. This behavior is consistent with the expected effect of R203 at the selected concentration within the recommended processing range. Conversely, even a minimal amount of Nexamite R202 causes a marked increase in MFI, indicating significant polymer fluidization.

Table 13. Melt flow index of the reprocessed formulations.

Formulations	PP REC 8.1	PP REC 8.2	PP REC 8.3
Melt Flow Index (g/10 min)	19.96 ± 0.4	8.62 ± 0.3	25.95 ± 0.5
Nexamite R202 (%)			0.04
Nexamite R203 (%)		4	4
ADK STAB A-611	0.5	0.2	0.2

Ref. [21] also shows that the degradation of recycled polypropylene produces persistent effects on the molecular structure and mechanical properties of the material, which cannot be fully recovered through simple conventional stabilization. This finding is consistent with the observations of the present study, where the addition of antioxidant alone (PP REC 8.1) does not allow significant recovery of performance, making a reactive rebuilding strategy necessary. Figure 9 shows the DSC curves for the PP REC 8.1, 8.2, and 8.3 formulations. The melting temperatures show limited differences of ±2–4 °C, with higher values for formulation 8.2, likely associated with molecular rebuilding and branching effects induced by Nexamite R203. This result is confirmed by the crystallinity values reported in Table 14. For PP REC 8.3, there is a decrease in melting temperature and crystallinity due to the 0.04% peroxide, which also slightly reduces the material's properties.

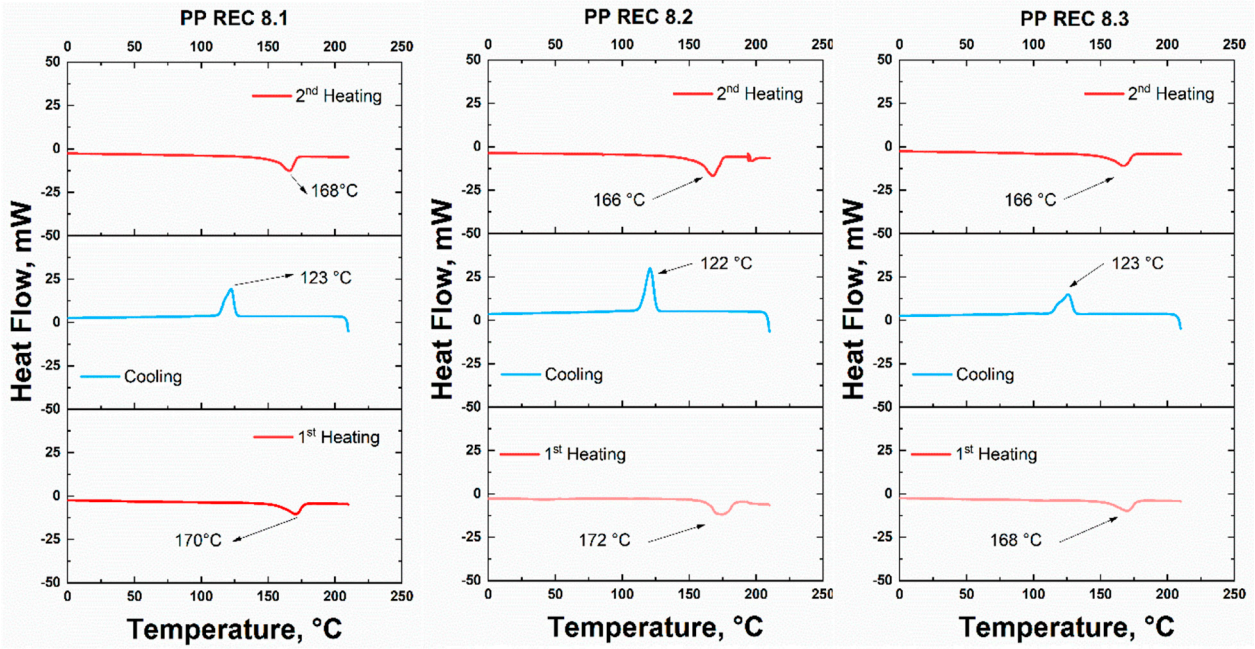


Figure 9. DSC curves of the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

Table 14. Degree of crystallinity, melting temperature, and crystallization temperature of the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

	PP REC 8.1	PP REC 8.2	PP REC 8.3
Crystallinity degree	$X_{CL} = 0.33$	$X_{CL} = 0.36$	$X_{CL} = 0.32$
Melting temperature (1st heating)	170 °C	172 °C	168 °C
Crystallization temperature.	123 °C	122 °C	123 °C
Melting temperature (2nd heating)	168 °C	166 °C	166 °C

The behavior is typical of polypropylene, exhibiting shear-thinning, with a strong decrease in viscosity as the shear rate increases. In the PP REC 8.2 formulation, the viscosity at low shear rates is higher compared to 8.1, and the curve shows a steeper initial slope, indicating more pronounced shear-thinning behavior and greater temperature sensitivity. This trend suggests the presence of possible molecular rebuilding, branching, or localized thermal stabilization induced by the additive. Conversely, the PP REC 8.3 formulation shows lower viscosities and thus higher melt fluidity, fully consistent with the MFI test results. These findings are confirmed by the values reported in Table 15, which show the viscosities at a shear rate of 1000 1/s at 180 °C, 190 °C, and 200 °C, respectively. Figure 10 shows the shear viscosity curves of the reprocessed formulations PP REC 8.1, PP REC 8.2, and PP REC 8.3.

Table 15. Viscosity values at a shear rate of 1000 s^{-1}.

Sample	180 °C (Pa·s)	190 °C (Pa·s)	200 °C (Pa·s)
PP REC 8.1	52	40	32
PP REC 8.2	66	53	41
PP REC 8.3	58	42	36

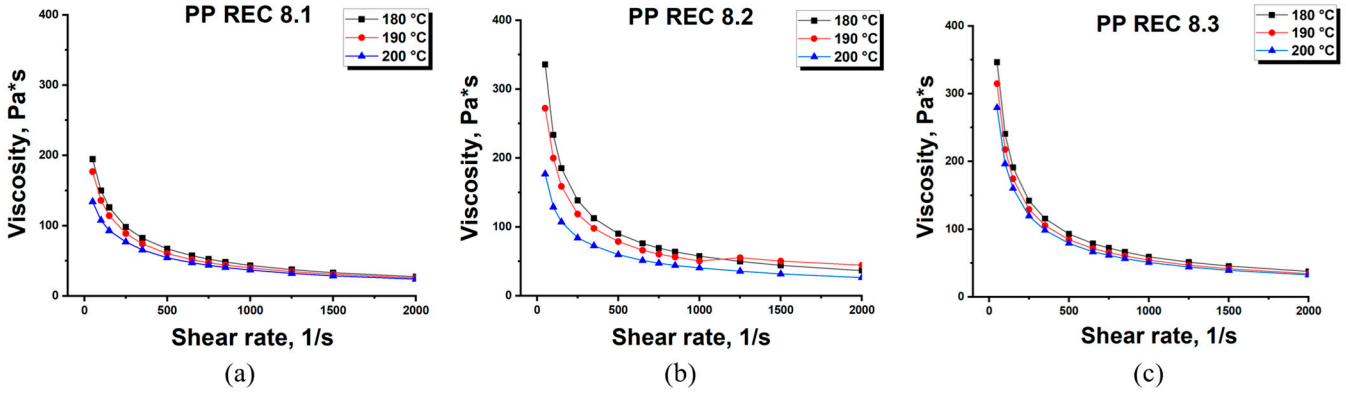


Figure 10. Shear viscosity curves of (**a**) PP REC 8.1, (**b**) PP REC 8.2, and (**c**) PP REC 8.3 formulations.

Figure 11 shows the FTIR spectra for each PP REC 8.1, 8.2, and 8.3 formulation. All formulations exhibit the typical polypropylene doublet. The polymer's basic chemical structure remains unchanged. In the 1400–1500 cm^{-1} region, the peaks are almost overlapping, showing no significant changes in the polymer backbone structure. In the 1000–1200 cm^{-1} region, the bands are aligned. No new functional groups are introduced by the upgrading procedures. Spectra containing Nexamite R203 and R202 show additional peaks in the 1500–900 cm^{-1} region. Table 16 reports the carbonyl index values for the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations. The carbonyl index values indicate that PP REC 8.2 has the lowest degree of oxidation (CI = 0.20), whereas PP REC 8.1 and PP REC 8.3 show significantly higher carbonyl content (CI = 0.36 and 0.38), indicating a greater level of oxidative degradation of the recycled material.

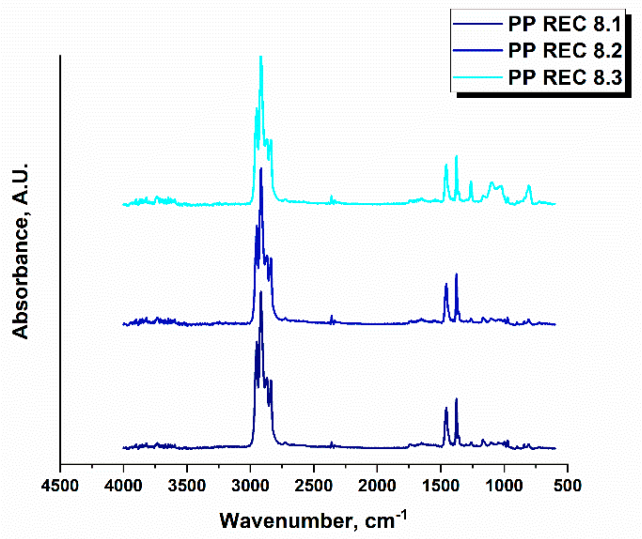


Figure 11. FTIR spectra of the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

Table 16. Carbonyl index for the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

Formulations	Carbonyl Index
PP REC 8.1	0.36
PP REC 8.2	0.20
PP REC 8.3	0.38

4.2. Tensile, Izod and HDT Test

Figure 12 shows the stress–strain curves of the reprocessed formulations PP REC 8.1, PP REC 8.2, and PP REC 8.3. Compared to the PP REC 8 formulation, there is a slight increase in stress, with average maximum values of 21 MPa. For strain, the values are more differentiated, with a notable increase for PP REC 8.2, showing average values of 78% as reported in Table 17. This indicates that the addition of Nexamite R203 does not significantly affect the stress variation, while strain increases considerably. For the PP REC 8.3 formulation, the values are comparable to those of PP REC 8 analyzed previously. The PP REC 8.2 material demonstrates the most favorable mechanical behavior, with high ductility ($\varepsilon_r \approx 60$–95%) and excellent reproducibility among specimens, indicating greater structural homogeneity.

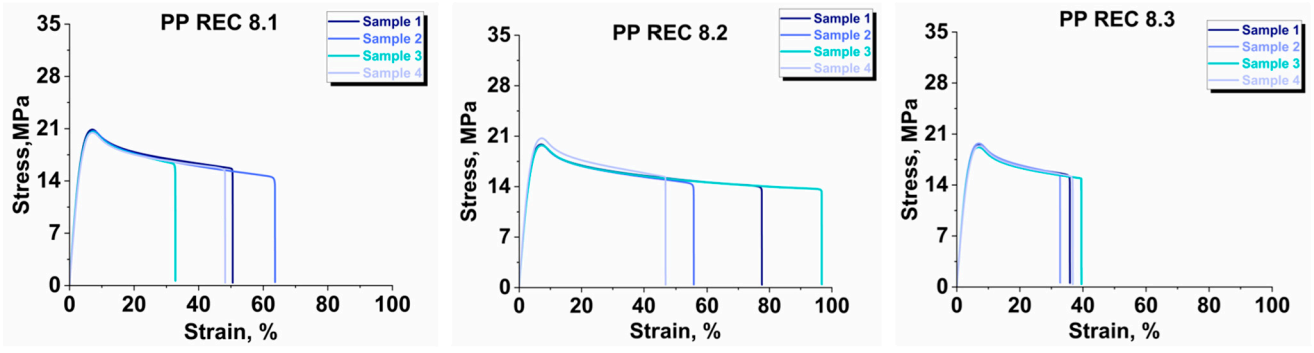


Figure 12. Stress–strain curves of the (**a**) PP REC 8.1, (**b**) PP REC 8.2, and (**c**) PP REC 8.3 formulations.

Table 17. Strain and stress values for the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

Mechanical Properties	PP REC 8.1	PP REC 8.2	PP REC 8.3
Strain (%)	48 ± 13.5	78 ± 21.0	35 ± 4.2
Stress (MPa)	20 ± 0.5	21 ± 0.6	18.5 ± 0.5

Figure 13 and Table 18 show, respectively, the box plot and the average values of energy impact. The average impact energy progressively increases from PP REC 8.1 to PP REC 8.3. PP REC 8.1 exhibits the lowest average value (approximately 3.1 kJ/m^2) and the greatest data dispersion, indicating marked material heterogeneity. PP REC 8.2 shows the highest average value (approximately 4.36 kJ/m^2) with reduced dispersion compared to 8.1, suggesting better structural uniformity. PP REC 8.3 shows an intermediate average value (approximately 3.75 kJ/m^2) and a very narrow data distribution, indicative of a more uniform material with greater energy dissipation capacity upon impact. Comparison with the tensile tests indicates that PP REC 8.3, despite exhibiting slightly reduced quasi-static ductility, has superior impact toughness, suggesting a morphology or microstructure favorable for energy dissipation at high deformation rates.

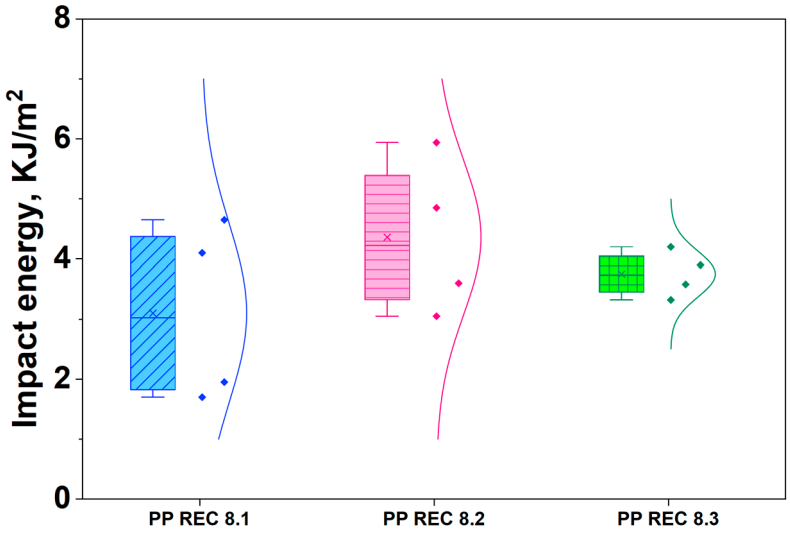


Figure 13. Box plot of the impact energy distribution for the PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

Table 18. Impact energy values for PP REC 8.1, PP REC 8.2, and PP REC 8.3 formulations.

Formulations	PP REC 8.1	PP REC 8.2	PP REC 8.3
Impact energy (kJ/m^2)	3.10 ± 1.55	4.36 ± 1.15	3.75 ± 0.36

The characteristic temperature reported in Table 19 is high and comparable for all formulations, with values ranging from approximately 79 to 83 °C. PP REC 8.2 shows the highest value (82.6 °C), followed by PP REC 8.1 (80.6 °C), while PP REC 8.3 has a slightly lower value (79.1 °C). The differences between the samples are small, indicating that the three formulations possess similar thermal stability. The slight increase observed for PP REC 8.2 suggests a thermally more stable microstructure, potentially related to a higher degree of crystallinity or a more favorable molecular weight distribution. Overall, the results indicate that compositional variations between 8.1, 8.2, and 8.3 do not substantially affect this thermal property, which remains essentially constant within the PP REC 8 series.

Table 19. Heat deflection temperature values.

Formulations	Temperature (°C)
PP REC 8.1	80.6
PP REC 8.2	82.6
PP REC 8.3	79.1

5. Conclusions

This study investigated the effect of controlled degradation and subsequent reactive reprocessing of polypropylene using Nexamite reactive additives, in order to modulate the rheological, thermal, and mechanical properties of recycled polypropylene. The addition of Nexamite R202 at 0.5%, 1%, and 2% induced progressive polymer chain scission, as evidenced by the increase in melt flow index, reduction in melt viscosity, decrease in crystallinity, and increase in carbonyl index. These changes resulted in a deterioration in mechanical performance for formulations with higher peroxide content (2%), particularly PP REC 9, which showed reduced strain, stress, impact energy, and thermal stability under load. Among the degraded formulations, PP REC 8 (1% Nexamite R202) represented the best compromise between melt fluidity and mechanical performance, combining good resistance under both static and dynamic loading. Subsequent reprocessing with Nexamite R203 and an antioxidant package enabled a significant recovery of the degraded material's performance. In particular, PP REC 8.2 showed increased melt viscosity, slight stabilization of thermal properties, and marked improvement in mechanical performance, with tensile stress of 21 MPa compared with 19.5 MPa for the starting material PP REC 8 (increase of approximately 7.7%), together with a substantial increase in elongation at break. The results demonstrate that controlled degradation followed by targeted reactive reprocessing is an effective strategy to tune processability and recover the performance of recycled polypropylene. This two-step approach combines the flow advantages generated during controlled degradation with the mechanical and thermal recovery achieved during reactive upgrading. Rather than restoring the material to its original state, the treatment enables a new balance between melt processability, ductility, and mechanical performance, offering promising opportunities for higher-value recycled PP applications. A limitation of the present work is the absence of direct molecular weight distribution data (e.g., GPC) and long-term durability testing. Future work will include GPC characterization, accelerated aging tests, and optimization of additive concentration to further validate the industrial potential of the proposed strategy.

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