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MECHANISMS OF POLYOLEFIN (PP AND PE) DEGRADATION AND COMPARATIVE CHARACTERISTICS OF PRIMARY AND SECONDARY POLYOLEFINS

Abstract: *This review systematizes recent open-access data on the degradation of polyolefins, with emphasis on polypropylene (PP) and polyethylene (PE) and on the differences between virgin and secondary materials during ageing, processing, and reprocessing. The analysis covers photo-oxidative, thermo-oxidative, and thermomechanical pathways and relates them to changes in molecular architecture, molecular weight, melt rheology, thermal stability, and mechanical performance. It is shown that polypropylene degrades predominantly through chain scission because tertiary carbon atoms are highly reactive; as a result, the average molecular weight decreases and the melt flow index usually increases. Polyethylene exhibits a more complex scenario in which chain scission competes with branching and partial cross-linking, causing non-linear viscosity behavior and a higher risk of embrittlement at advanced oxidation stages. Comparative data indicate that recycled PE and PP generally lose tensile strength, impact resistance, and elongation at break relative to virgin grades, although properly selected blend composition, antioxidant stabilization, and rheology-based quality control can significantly improve recyclate performance. The practical conclusion is that recycled*

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polyolefins should be evaluated using mechanism-based diagnostics, including Fourier-transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC), size-exclusion chromatography (SEC), oxidation induction temperature (OIT), and rheological criteria, rather than relying on a single processing parameter.

Key words: *polyolefins, photo-oxidation, thermo-oxidation, mechanical recycling, secondary polymers, stabilization.*

Introduction

Polyethylene and polypropylene constitute the bulk of commercial plastics and at the same time dominate the stream of polymer waste; therefore, the issue of polyolefin degradation is directly related to the quality of mechanical recycling and to the realism of the circular economy [1-5]. This review focuses on the degradation mechanisms of polypropylene (PP) and polyethylene (PE), methods for diagnosing degradation, and the differences in the behavior of virgin and recycled materials [1,2, 6-8].

The aim of this work is to systematize the mechanisms of photo-oxidative, thermo-oxidative, and thermomechanical degradation of PP and PE and to show how these processes change the structure, molecular weight, rheological behavior, and mechanical properties of recycled polyolefins. The working hypothesis is that the difference between virgin and recycled polyolefins is determined not only by the degree of oxidation but also by the predominant type of molecular response: for PP, this is mainly chain scission, whereas for PE it is a combination of chain scission, macromolecular chain branching, and limited cross-linking [2, 7-10].

The relevance of the topic is reinforced by the fact that recycled materials are increasingly viewed not as a low-grade substitute for virgin polymers but as an independent class of engineering feedstock [11-13]. Therefore, for the practical use of recycled PP and PE, it is not enough to measure only strength or melt flow index; it is necessary to relate service properties to the degradation mechanism, processing history, and residual stabilization of the material [14-16].

Unlike reviews focused primarily on individual degradation mechanisms, stabilization strategies, or specific recycling technologies, this review considers polypropylene and polyethylene comparatively, with particular attention to the relationship between degradation mechanisms, changes in molecular architecture, diagnostic parameters, and the resulting properties of virgin and recycled materials. The review brings together data on photo-oxidative, thermo-oxidative, and thermomechanical degradation and relates these changes to approaches for the quality assessment and property recovery of recyclates. Thus, the focus is not only on describing degradation phenomena, but also on providing a mechanism-based framework for interpreting differences between virgin and secondary polyolefins and for selecting appropriate diagnostic and processing strategies.

Materials and methods

The work was carried out as a targeted analytical review. The review mainly included open publications from 2020-2025 devoted to the degradation of PP and PE during ageing, processing, and reprocessing. Priority was given to studies that simultaneously presented: (1) the degradation mechanism of PP and/or PE; (2) quantitative indicators of property changes; (3) reproducible methods of instrumental control; and (4) results applicable to comparing virgin and recycled polyolefins. In addition to recent open-access studies, a limited set of earlier foundational works and review papers was included where they were necessary to explain the core mechanisms of degradation, restabilization, and recyclate quality assessment [2-4, 8, 12, 16].

The analysis included data on photo-oxidation, thermo-oxidation, thermomechanical degradation, antioxidant stabilization, and rheological criteria for the quality of recycled feedstock [3, 8, 14-18]. When interpreting the results, the polymer type, ageing or processing conditions, the direction of changes in molecular architecture, and the corresponding response of mechanical and rheological characteristics were compared.

To compare the data, the following groups of indicators were used: chemical markers of oxidation (carbonyl and hydroxyl groups by Fourier-transform infrared spectroscopy (FTIR)), molecular-weight characteristics and indirect rheological indicators (melt flow index (MFI), complex viscosity, and the degradation value (V_{deg}) parameter), thermal indicators (oxidation temperature and oxidation induction temperature (OIT)), as well as standard mechanical properties, including tensile strength, elongation at break, impact strength, and flexural strength [14-16, 19, 20]. The illustrations in the review are placed in the sections where they strengthen the interpretation of the mechanism or confirm quantitative conclusions.

Results and discussion

Structural prerequisites for the differences between PP and PE

The key difference between PP and PE is determined by chain structure. In polypropylene, the presence of a tertiary carbon atom facilitates hydrogen abstraction and accelerates the formation of macroradicals; therefore, when heated and in the presence of oxygen, PP tends toward β-scission and a decrease in average molecular weight [6-8, 21]. For polyethylene, the situation is less unambiguous: depending on the type of PE, temperature, oxygen availability, and material history, chain scission, chain branching, and partial cross-linking occur in parallel, and for high-density polyethylene (HDPE) an increase in effective molecular weight is often observed owing to branching and microgel formation (Table 1) [7, 9, 22]. This difference explains why recycled PP usually shows an increase in melt flow, whereas recycled PE shows either a decrease or nonlinear behavior as oxidation develops [14, 18, 23-25].

Table 1 – Comparison of the predominant degradation mechanisms of PP and PE

Parameter	Polypropylene (PP)	Polyethylene (PE)
Most vulnerable structural unit	tertiary carbon atom and its adjacent C–H bonds	secondary C–H bonds; lower reactivity
Dominant response to processing	chain scission, decrease in average molecular weight, increase in MFI	chain scission + branching/cross-linking; viscosity may increase
Typical result of photo- and thermo-oxidation	increase in carbonyl index and decrease in ductility	increase in carbonyl index and crystallinity, accompanied by surface cracking
Risk for recycled material	loss of impact strength and elongation	unpredictable rheology, brittleness after prolonged ageing

Note – compiled based on data from [6-10, 21].

Photo-oxidation and thermo-oxidation of polyolefins

Photo-oxidative degradation does not begin with light absorption by an ideal hydrocarbon matrix, but with the activation of impurity chromophores and already formed oxygen-containing groups [17, 21, 26, 27]. For both polyolefins, the autocatalytic cycle is fundamental: radical formation, oxygen addition, hydroperoxide formation, and their decomposition with the initiation of new oxidation events (Figure 1) [6-8, 21]. In PP, this cycle more rapidly drives the system toward chain scission; in PE, it is more often accompanied by competing branching and cross-linking reactions [7-10]. Long-term thermal ageing of PP is further complicated by heterogeneity: local oxidation sites can spread through the material and through the gas phase, which reduces the predictability of secondary processing even at the same average chemical composition [6, 28, 29].

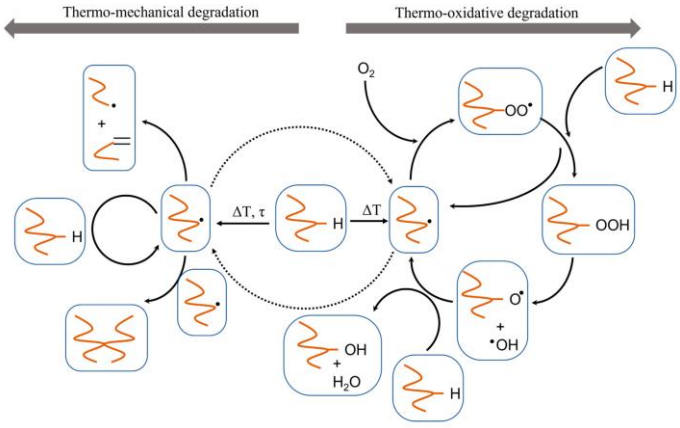


Figure 1 – General pathways of thermal and thermo-oxidative degradation of polymers; the scheme clearly illustrates the autocatalytic cycle common to PP and PE [1]

For polypropylene, long-term thermo-oxidative stability is especially sensitive to stabilization. According to the review by P. Gijsman and R. Fiorio, unstabilized PP at 80 °C degrades in about 32 days, whereas the introduction of 500 ppm of a phenolic antioxidant can increase the time to failure

to more than five years. This is important for recycled feedstock: the quality of the recyclate is determined not only by the degree of oxidation that has already occurred, but also by how properly the virgin grade was stabilized in its first life cycle [6, 16, 30].

For polyethylene, photo-oxidation often develops according to a staged scenario: the early stage is accompanied by a decrease in molecular weight; then crystallinity and the degree of structural fixation increase, which slows further oxidation but simultaneously increases brittleness [17, 27, 31]. In a study of Low-Density Polyethylene (LDPE) sheet degradation after 8 weeks of UV ageing, the molecular weight of the initial material decreased from 2.09×10^5 to 1.45×10^4 Da, and the use of pro-oxidants additionally accelerated degradation and increased crystallinity; therefore, the mere presence of oxygen-containing groups does not in itself imply improved processability [17].

Thermomechanical degradation during processing and reprocessing

During extrusion, injection moulding, and other processing operations, polyolefins are simultaneously subjected to thermal, mechanical, and oxidative stress [1, 4, 8, 18, 32]. For PP, the combination of shear and temperature usually leads to repeated chain scission and increased flowability [10, 18, 24, 33-35]. In the work of E. Esmizadeh and co-authors, reprocessing simulation showed that after only five mixing cycles the steady-state melt torque decreased by about 35%, while the melt flow index increased by more than 400% [6]. This is direct evidence of strong degradation of macromolecules and a reduction in average chain length [10, 18, 24, 36].

For PE, degradation during repeated processing is often less linear [9, 22, 25, 37]. In film systems containing virgin LLDPE, post-industrial rLLDPE, and post-consumer rPE, repeated cycles lead to deterioration of tensile strength and tear resistance, but some properties are retained due to structural rearrangement, changes in branching, and partial cross-linking [38-40]. Therefore, interpreting changes in the flowability of recycled PE without FTIR, differential scanning calorimetry (DSC), or size-exclusion chromatography (SEC) is risky: the same MFI may correspond to different oxidation levels and different residual strength [12, 14, 20, 25].

Comparison of virgin and recycled polyolefins based on quantitative data

The clearest comparison is obtained by comparing virgin and recycled materials of the same chemical class [12, 14, 39]. In the work of W. Zhang and co-authors, the melt flow index for PE decreased from 2.191 to 0.752 g/10 min after ageing, i.e., by 65.68%, whereas for PP it increased from 8.254 to 11.486 g/10 min, or by 39.16% (Table 2) (Figure 2) [23]. This multidirectional change confirms that chain scission dominates in recycled PP, whereas for recycled PE the later stages of degradation involving branching and cross-linking begin to play a substantial role [7, 9, 10, 23, 41].

Recent results further indicate that PP degradation during mechanical recycling can be related to its previous thermo-oxidative aging during service, as thermo-mechanical processing may induce additional changes in mechanical and thermal properties and affect the material's potential for a second life cycle [42].

Table 2 – Mechanical properties of virgin and recycled PE and PP

Property	vPE	rPE	vPP	rPP
Tensile strength, MPa	38.57	24.12	31.26	27.90
Elongation at break, %	168.44	44.23	107.45	67.52
Notched impact strength, kJ/m²	8.16	4.05	7.18	5.19
Flexural strength, MPa	32.52	22.63	29.43	21.43

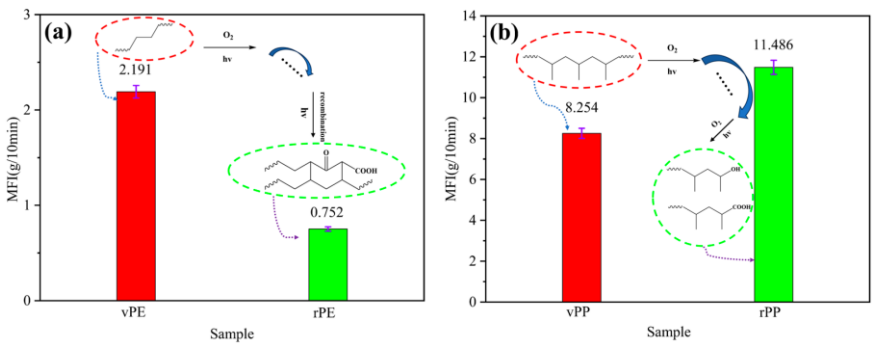


Figure 2 – Comparison of MFI (melt flow index) for virgin and recycled PE and PP [23]

The decrease in mechanical properties is also material-specific [23, 25, 39, 43]. For rPE, elongation at break decreased by 73.74% compared with the virgin material, whereas for rPP the decrease was 37.16%. In other words, recycled PE loses deformability more severely, which is consistent with the accumulation of oxidized groups, surface cracking, and the subsequent stiffening/embrittlement of the supramolecular structure [9, 17, 27]. At the same time, recycled PP loses molecular weight more rapidly and flows more readily in the melt, but under moderate degradation it still retains part of its load-bearing capacity [18, 24, 44].

For rPE/rPP blends, the most successful composition was 6:4 by weight: impact strength reached 7.48 kJ/m² and elongation at break reached 98.14% (Figure 3) [23]. This is important technologically because it shows that recycled polyolefins should not always be assessed separately [24, 35]. With the correct component ratio and the presence of oxidized functional groups, interfacial interactions are possible that increase the viscoelastic response and impact resistance [23, 45-48].

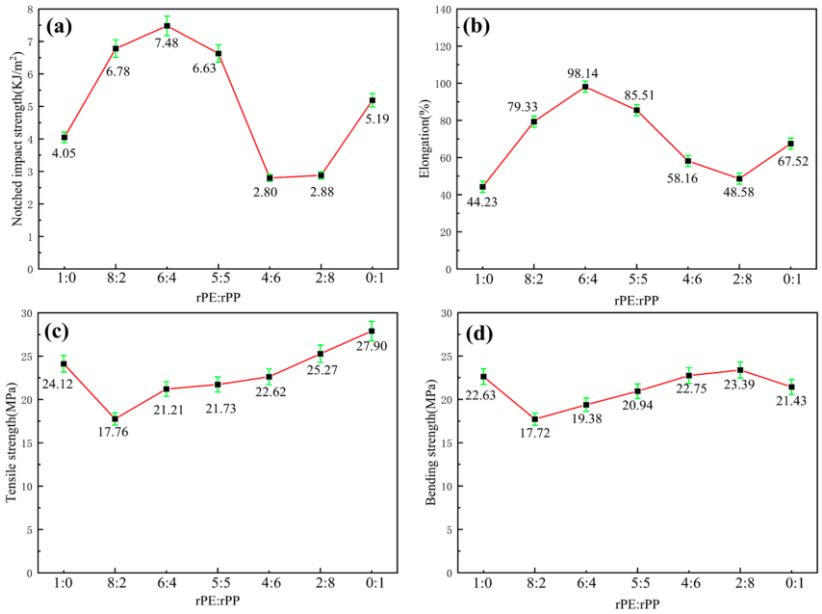


Figure 3 – Mechanical properties of rPE/rPP blends at different component ratios; maximum impact strength and elongation are achieved at 6:4 [23]

Antioxidant stabilization and quality-control methods for recycled feedstock

From the standpoint of practical recycling, not only the damage that has already occurred is important, but also the residual ability of the material to resist new oxidation [15, 16, 30, 49]. For recycled PP, this is especially clearly shown in the work of N. Knoben and co-authors: without the addition of antioxidants, OIT decreased sequentially from 225 to 198°C, whereas in a closed-loop cycle with additives an increase in OIT to 257°C was observed. At the same time, by the fifth cycle the content of Irgafos 168 increased to about 4170 ppm and Irganox 1010 to about 2050 ppm, indicating a risk of additive accumulation [19]. Consequently, stabilization improves processability, but in a closed loop it requires quantitative control, especially for packaging applications [16, 19, 30, 50].

Classical indicators such as MFI or a single viscosity point do not always make it possible to distinguish beneficial branching from critical degradation [12, 14, 20].

Therefore, the rheological parameter V_{deg}, proposed for assessing the quality of recycled HDPE, is promising. This indicator is calculated from changes in the shape of Van Gurp-Palmen curves and is linked to the degree of long-chain branching [14]. For HDPE, the change in complex viscosity in an inert atmosphere was about 14% at 10 rad/s, whereas in air the effect was about five times stronger at 0.1 rad/s (Figure 4) [14]. Higher V_{deg} values and an earlier plateau indicate a greater extent and/or faster development of structural rearrangement during reprocessing, which can be associated with lower recyclate quality [12, 14, 20, 25].

$$V_{\text{deg}} = \lim_{0.31 \le \omega \le 3.16} \frac{\partial \delta}{\partial |G^*|}$$

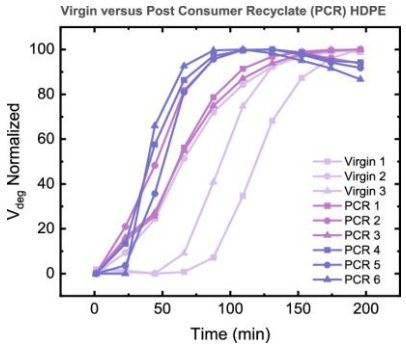


Figure 4 – Example of using the V_{deg} parameter for rapid assessment of recycled HDPE quality [14]

Additional data from recent studies on recycled PP

Recent publications supplement the main logic of the article primarily in the part where it is necessary to show the practical difference between acceptable processability of recycled PP and its residual resistance to further ageing [11, 15, 16, 30]. In the study on virgin/recycled polypropylene blends, it is shown that an increase in recycled content is accompanied by an increase in melt flow index, whereas the oxidation induction time decreases much faster [11]. Consequently, even a material that is technologically convenient in terms of flowability may have a limited reserve of antioxidant stability, which is important to take into account during re-extrusion and when choosing the fraction of rPP in a composition [11, 15, 16, 19, 30] (Table 3).

An additional applied aspect is revealed in the study devoted to post-consumer recycled polypropylene for automotive applications [51]. The authors showed that with proper formulation and feedstock preparation, recycled PP can retain acceptable thermal and mechanical stability as well as resistance to UV ageing [16, 51-54]. At the same time, the most sensitive characteristics remain low-temperature impact strength and, partially, flexural stiffness, i.e., precisely those properties that limit the use of recycled PP in the most highly loaded and cold-resistant parts [44, 51, 52] (Table 3).

Table 3 – Recent data supplementing the comparative characteristics of virgin and recycled PP

Source	Key observations	Significance for the article
[11]	As the fraction of recycled PP increases, MFI rises and oxidative stability decreases; already at 25% rPP, the oxidation induction time decreases by about 71%.	Confirms that processability and reserve oxidative stability should be assessed separately.
[51]	Post-consumer recycled polypropylene (PCR-PP) compounds for automotive applications retain acceptable thermal and mechanical properties and good UV resistance, but are inferior in low-temperature impact strength.	Shows the limits of applicability of recycled PP in technical products.

Practical conclusions for comparing virgin and recycled polyolefins

Comparison of contemporary data makes it possible to formulate several practical conclusions. First, recycled PP more often needs molecular-weight control and antioxidant replenishment, because chain scission quickly moves the material into the region of high flowability and loss of impact resistance [11, 15, 18, 19]. Second, recycled PE must be assessed more comprehensively: a decrease in MFI does not mean improvement, but may reflect a late stage of oxidation, branching, and a latent tendency toward brittle failure [9, 14, 17, 25]. Third, for mixed PE/PP streams, formulation approaches are promising in which degradation-induced functional groups are used not only as a defect but also as a source of interfacial interaction in compatible compositions [23, 45-48].

Finally, the difference between virgin and recycled polyolefins should be considered not as a binary opposition of “high-quality/low-quality material”, but as a continuum of structural state [12, 14, 16]. Given FTIR, DSC, SEC, and rheological data, recycled polyolefins can be classified quite accurately by degree of degradation and appropriate target applications can be selected for them – from less demanding film products to modified blends and composite materials [12, 14, 20, 39]. This classification becomes even more reliable when rheological screening is combined with additive-assisted property recovery strategies for recycled PP [15, 16, 30].

Conclusion

The analysis carried out showed that for the topic of PP and PE degradation, the three interrelated blocks of greatest importance are: the mechanism of formation of oxygen-containing defects, the change in molecular architecture during processing, and the resulting loss of mechanical properties of recycled materials. For polypropylene, chain scission associated with the reactivity of the tertiary carbon atom dominates; for polyethylene, a combination of chain scission, branching, and partial cross-linking is characteristic. This is why recycled PP more often demonstrates an increase in melt flow, whereas recycled PE shows more complex and sometimes opposite rheological behavior.

Quantitative data from recent open sources confirm that degradation worsens the strength, impact resistance, and especially the deformability of recycled polyolefins; however, these losses do not preclude the engineering use of recyclate. The most promising approaches are targeted blending of rPE and rPP, management of antioxidant stabilization, and implementation of rapid quality-control methods based on OIT and rheological criteria. The transition presented in this work from analysis of degradation mechanisms to the formation of an applied scheme of “degradation mechanism – diagnostic parameters – technological solutions” opens possibilities for targeted regulation of the properties of recycled polyolefins and for the development of methods for their rapid monitoring under processing conditions.

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ПОЛИОЛЕФИНДЕРДІҢ (ПП ЖӘНЕ ПЭ) ДЕСТРУКЦИЯ МЕХАНИЗМДЕРІ ЖӘНЕ БАСТАПҚЫ МЕН ҚАЙТАЛАМА ПОЛИОЛЕФИНДЕРДІҢ САЛЫСТЫРМАЛЫ СИПАТТАМАСЫ

Бұл шолу жұмысы полиолефиндердің деструкциясы туралы соңғы ашық қолжетімді деректерді жүйелейді және полипропилен (ПП) мен полиэтиленнің (ПЭ), сондай-ақ бастапқы және қайталама материалдардың қартаю, өңдеу және қайта өңдеу кезіндегі айырмашылықтарына ерекше назар аударады. Талдауда фото-тотығу, термо-тотығу және термомеханикалық деструкция жолдары қарастырылып, олардың молекулалық архитектураға, молекулалық массаға, балқыма реологиясына, жылулық тұрақтылыққа және механикалық қасиеттерге әсері салыстырылды. Көрсетілгендей, полипропилен негізінен тізбектің үзілуі арқылы ыдырайды, өйткені үшінші реттік көміртек атомдары жоғары реакциялық қабілетке ие; соның салдарынан орташа молекулалық масса төмендеп, балқыма ағынының индексі көбіне артады. Полиэтиленде үзілу процестері тармақталумен және ішінара торланумен қатар жүреді, сондықтан тұтқырлықтың өзгерісі сызықтық емес сипат алып, тотығудың терең сатысында морттану қаупі өседі. Салыстырмалы деректер қайталама PE мен PP-де созылу беріктігі, соққыға төзімділік және үзілу кезіндегі салыстырмалы ұзару әдетте бастапқы материалдарға қарағанда төмендейтінін көрсетті. Дегенмен қоспа құрамын дұрыс таңдау, антиоксиданттық тұрақтандыру және реологияға негізделген сапаны бақылау қайта өңделген материалдардың қолданылу мүмкіндігін едәуір жақсартады. Практикалық тұрғыдан екіншілік полиолефиндерді бір ғана технологиялық көрсеткішке сүйенбей, деградация механизмдеріне негізделген кешенді диагностика арқылы, оның

ішінде Фурье-түрлендіру инфрақызыл спектроскопиясы (FTIR), дифференциалды сканерлеуші калориметрия (DSC), эксклюзиялық хроматография (SEC), тотығу индукциясының температурасы (OIT) және реологиялық критерийлерді пайдалана отырып бағалау қажет.

Түйін сөздер: *полиолефиндер, фото-тотығу, термо-тотығу, механикалық қайта өңдеу, қайталама полимерлер, тұрақтандыру.*

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МЕХАНИЗМЫ ДЕСТРУКЦИИ ПОЛИОЛЕФИНОВ (ПП И ПЭ) И СРАВНИТЕЛЬНАЯ ХАРАКТЕРИСТИКА ПЕРВИЧНЫХ И ВТОРИЧНЫХ ПОЛИОЛЕФИНОВ

В обзоре систематизированы современные открытые данные о деструкции полиолефинов с акцентом на полипропилен (ПП) и полиэтилен (ПЭ), а также на различия между первичными и вторичными материалами при старении, переработке и повторной переработке. Рассмотрены фотоокислительные, термоокислительные и термомеханические пути деградации и показано, как они отражаются на молекулярной архитектуре, молекулярной массе, реологических свойствах расплава, термической стабильности и механических характеристиках. Показано, что полипропилен преимущественно деградирует по механизму разрыва цепи вследствие высокой реакционной способности третичных атомов углерода; поэтому средняя молекулярная масса снижается, а показатель текучести расплава обычно возрастает. Для полиэтилена характерен более сложный сценарий, при котором разрыв цепи сочетается с разветвлением и частичным сшиванием, что приводит к нелинейному изменению вязкости и повышенному риску охрупчивания на поздних стадиях окисления. Сопоставление количественных данных показывает, что вторичные PE и PP, как правило, уступают первичным материалам по прочности при растяжении, ударной вязкости и относительному удлинению при разрыве. Вместе с тем правильно подобранный состав смесей, антиоксидантная стабилизация и реологический контроль качества способны существенно расширить область применения вторичных полиолефинов. Практический вывод заключается в том, что вторичные полиолефины следует оценивать с использованием диагностики, основанной на механизмах деградации, включая инфракрасную Фурье-спектроскопию (FTIR), дифференциальную сканирующую калориметрию (DSC), эксклюзионную хроматографию (SEC), температура окислительной индукции (OIT) и реологические параметры, а не полагаться на один единственный параметр переработки.

Ключевые слова: *полиолефины, фотоокисление, термоокисление, механическая переработка, вторичные полимеры, стабилизация.*

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ФЛОКУЛЯЦИЯ ГИДРОСУСПЕНЗИИ БЕНТОНИТОВОЙ ГЛИНЫ, ИСПОЛЬЗУЕМОЙ В ОЧИСТКЕ ШАХТНОЙ ВОДЫ ГОРНОРУДНОЙ ПРОМЫШЛЕННОСТИ, ПОЛИАКРИЛАМИДНЫМИ ПОЛИЭЛЕКТРОЛИТАМИ

Аннотация: *Восточно-Казахстанская область обладает значительными запасами бентонитовых глин Таганского месторождения, которые характеризуются высокой сорбционной емкостью и способностью эффективно удалять ионы тяжёлых металлов из шахтных вод горнорудной промышленности до уровня предельно-допустимой концентрации рыбохозяйственного назначения.*

Однако применение бентонита сопровождается увеличением мутности очищаемой воды, что ограничивает его практическое использование в водоёмах. Для решения данной проблемы

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