

Article

An Efficient Method for Recycling Polypropylene with the Antioxidant Additive Propyl Gallate

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Abstract

In this study, a new approach to stabilizing polypropylene (PP) melts using the bio-based antioxidant propyl gallate (PG) is considered. Since PG is extensively used in the food and cosmetics fields and its low doses do not cause reactions in humans, producing PP melts with PG is an interesting approach. Furthermore, PG is already actively used in the processing of natural polymers, for example, in the NMMO process. It is shown that introducing up to 0.2 wt.% PG into the system is sufficient to significantly reduce the decrease in melt viscosity during repeated PP processing. After five processing cycles, the viscosity of systems with PG decreases by less than an order of magnitude, while for melts without the antioxidant the viscosity drops by almost three orders of magnitude to 10 Pa s. For melts with antioxidant additives, the crossover point position in the frequency dependences remains virtually unchanged, indicating the preservation of the system's elastic properties. Macrofibers were spun from the resulting melts, which can then be used for concrete reinforcement. For the spun fibers with PG, the strength decreased to 68.1 MPa after five passes, whereas for the PP fibers, the values did not exceed 31 MPa. The structure and properties of the fibers were studied using X-ray diffraction and IR spectroscopy, and contact angles were determined.

Keywords: polypropylene; propyl gallate; antioxidant; melt; rheology; recycling; macrofibers



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1. Introduction

Despite the growing pressure on fossil raw materials, synthetic polymers remain in high demand in various industries. Among thermoplastics, polypropylene (PP) is one of the three most produced and consumed polymers [1]. Along with other common polyolefins, PP has low density, excellent processability, inertness towards a number of solvents, good mechanical strength, and an acceptable cost [2,3]. Due to its properties, PP is used in the food industry for product packaging (flexible and rigid), as structural elements, in the production of household and medical goods, in the automotive industry (bumpers, interior

elements), and in the textile industry (non-woven materials, fibers) [4–7]. The annual production of PP amounts to hundreds of millions of tons, which underlines its key role in modern industry [8].

However, the rapid increase in production and use of polypropylene, especially in the short-lived product segment (e.g., disposable packaging), has led to the formation of a comparable waste stream [9]. According to [10], no more than a quarter of the synthesized PP was reused. The remaining PP in the form of products accumulates in special landfills and pollutes the environment. Within the framework of the traditional linear economic model (“create, use, throw away”), used polypropylene products have become a serious source of environmental pollution. Since the decomposition period of polypropylene is estimated at hundreds of years, there is a risk of contamination of natural ecosystems and a negative impact on flora and fauna [11,12]. This has created an irresolvable contradiction between the economic benefits of producing and using PP-based products and the environmental damage caused primarily by uncontrolled polymer emissions into the environment and insufficient capacity to produce secondary PP products. The development of new approaches to PP recycling opens up opportunities for effective closed-loop strategies for this polymer.

It is known that polymer waste management systems are based on three main approaches: incineration, landfill, and recycling. The first two are inconsistent with the principles of a circular economy, as they are associated with significant environmental risks. Without focusing on the issue of PP landfill, we note that this is a passive strategy that leads to the loss of valuable raw materials [13]. PP’s high inertness to liquid environments and external influences leads to the accumulation of the polymer over centuries [14]. Its degradation leads to the formation of microplastic particles and the leaching of dyes and special additives.

Thermal processing of PP (incineration and pyrolysis) allows for obtaining a number of valuable products from the polymer and energy. But, as it is known, various additives (antioxidants, dyes, rheological additives, flame retardants) are often introduced into PP melts, which affect the kinetics of pyrolysis and polymer combustion [15,16]. The use of pyrolysis catalysts allows for achieving the desired results (high carbon residue, lower pyrolysis onset temperature) [17]. Proposed thermal recycling methods for PP require capital investment in the construction of modern incineration plants and pyrolysis units. Therefore, thermal recycling methods for PP have not gained widespread acceptance.

Mechanical recycling of PP has a number of advantages over thermal recycling methods and storage of the polymer in landfills. It involves simpler stages and is more economically accessible. The main stages of mechanical recycling are collecting PP, cleaning it from mechanical and liquid contaminants, crushing the polymer, and producing granules, which are then used to produce molded products [18,19]. Mechanical recycling of PP allows us to significantly reduce the use of virgin polymer, and therefore the fossil raw materials from which the polymer is synthesized. The carbon footprint is also reduced by reducing greenhouse gas emissions into the atmosphere. The number of PP recycling cycles can reach 9–14 [20,21]. Thermoplastic recycling also allows for the development of recycling infrastructure and the creation of new high-tech jobs. Therefore, mechanical recycling is considered a key method for closing the life cycle of polymeric materials and transitioning from a linear to a cyclical model [22].

Since secondary PP is practically not used for forming food or medical packaging, most of it goes to the production of building materials. Forming reinforcing fibers for concrete matrices based on PP is a promising and in-demand area. The introduction of polymer fibers into the concrete matrix makes it possible to control the formation and propagation of microcracks in the early stages of concrete hardening [23,24]. Unlike steel

reinforcement, PP fibers are uniformly distributed throughout the volume of the mixture, do not increase the weight of the structure, and are not subject to corrosion [25]. It has been shown that adding one kg per cubic meter of PP fibers increases the strength by several tens of percent [26], and the compressive strength can increase by up to 16% [27]. Numerous studies [28,29] have shown that the introduction of PP fibers into solutions can significantly reduce the risk of cracking and increase the strength and plasticity of concrete.

Depending on the spinning conditions, it is possible to obtain fibers of varying thickness (micro- and macrofibers). Fibers with a thickness of several tens of microns are used for controlled plastic shrinkage of concrete, while macrofibers are used for structural reinforcement [30–32]. Compared to macrofibers, microfibers do not significantly affect the mechanical properties (strength and rigidity) of concrete [33,34]. Therefore, the production of PP macrofibers is of primary interest for the formation of more durable concrete structures, especially in remote regions (e.g., in Kazakhstan) with low population density, where the transportation of building materials is difficult due to high costs.

The key task is to distribute the fibers uniformly throughout the volume of the concrete mixture, and the fibers should not have a pronounced orientation in the matrix. Schematically, the reinforcement principle can be represented as a set of randomly arranged monolithic strands of a given length that do not form agglomerates and interact with each other, creating a sparse mesh structure (Figure 1).

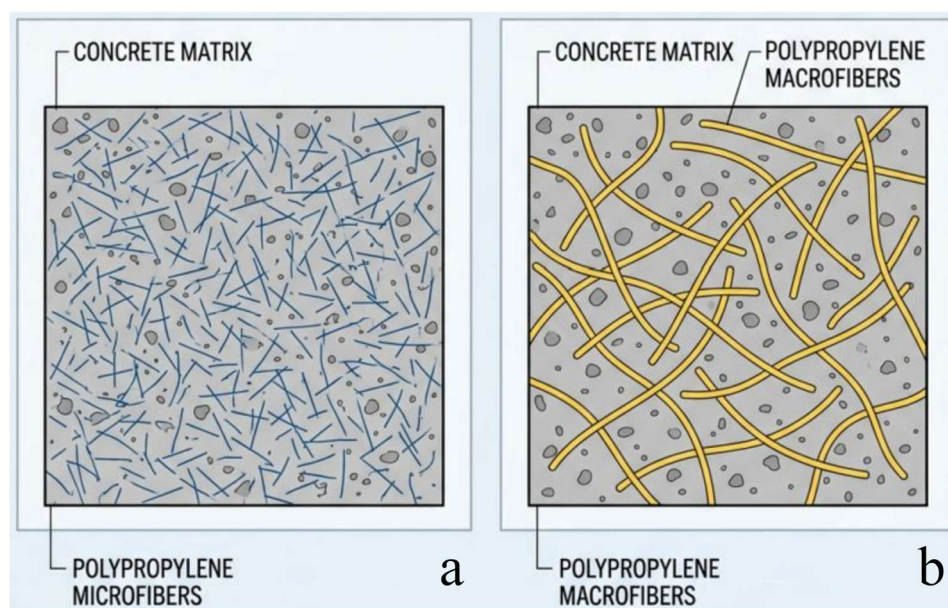


Figure 1. Schematic representation of concrete filled with micro- (a) and macrofibers (b).

However, mechanical recycling of PP is not a straightforward solution. It is known that many thermoplastics undergo degradation when exposed to high temperatures and high-pressure shear deformation (processing in an extruder) [35,36]. Thermomechanical and thermooxidative effects lead to changes in polymer color, a decrease in its molecular weight, a drop in viscosity and melt elasticity, and instability in fiber formation [37–39]. The mechanisms of chain scission and molecular weight reduction in PP are described in detail in the literature, so we will not dwell on them. However, methods for eliminating PP degradation and improving the performance properties of the molded materials are of great interest.

Rheological characteristics are the most sensitive to degradation. A decrease in melt viscosity and elasticity negatively impacts processing stability, complicates process control, and can impair the appearance and dimensional stability of finished products [40,41].

It is known from the literature that the introduction of viscosity stabilizers (antioxidants) into melts helps to mitigate the decrease in molecular weight and maintain the mechanical properties of the molded materials [42,43]. Traditionally, synthetic phenolic antioxidants and phosphites are used for this purpose [44]. However, growing demands for environmental friendliness and safety of materials are stimulating the search for alternative, potentially “greener” stabilizers that can be obtained from renewable raw materials or have an improved safety profile.

Natural antioxidants such as Vitamin E and others have been described previously [45,46]. Propyl gallate (PG) may be another interesting antioxidant additive. This additive is actively used in the food industry (additive E310), for the production of cellulose spinning solutions, etc. [47]. The antioxidant properties of the ester of gallic (3,4,5-trihydroxybenzoic) acid are due to the presence of phenolic hydroxyl groups in its molecule [48]. These groups are capable of effectively interrupting oxidation chain reactions by donating a hydrogen atom and transforming into stable radicals, which forms the basis of their antiradical action [49]. A similar mechanism is common to many industrial synthetic antioxidants, which suggests the potential effectiveness of propyl gallate in stabilizing polymer matrices during thermooxidative degradation, and we assume that PG will also exhibit antioxidant properties toward PP.

A key advantage of propyl gallate is its origin: it can be obtained from natural sources or by biosynthesis, which is consistent with current trends in renewable chemistry and the creation of safer materials [50]. Therefore, the aim of this study was to obtain PP melts with PG additives, investigate the rheological and morphological properties of the system, evaluate the moldability of the melts and the production of macrofibers from them, as well as determine the structural features of the resulting fibers and their mechanical properties.

2. Materials and Methods

To form the fiber, polypropylene homopolymer Balen 01030 (Ufaorgsintez, Birskiy trakt, Ufa, Russia), suitable for extrusion, was used. Melt index: 3.0 g/10 min (230 °C/2.16 kg). Density: 0.9 g/cm³. Melting point: ~165 °C.

Propyl gallate powder (Sigma Aldrich, St. Louis, MO, USA) was used as an antioxidant. A total of 0.2 wt.% PG was added to the PP during each recycling cycle.

Virgin PP and polymer that had undergone multiple recycling (simulation of recycling) were used in this study.

2.1. Rheology

An MCR 702 rheometer (Anton Paar GmbH, Graz, Austria) with a cone-plane geometry of 20 mm, 1° was used to evaluate the viscous and elastic properties of melts under continuous deformation in the shear rate range $\dot{\gamma}$ from 10^{−3} to 10³ s^{−1}.

Dynamic tests were conducted in the frequency range (ω) from 0.1 to 100 Hz at a constant stress of 10 Pa. The tests were conducted at a temperature of 185 °C.

2.2. Formation of Fibers

To obtain macrofibers, 0.2 wt.% PG was added to PP and mixed in a laboratory mixer at room temperature for 15 min. The resulting mechanical mixture was then fed into a twin-screw extruder (HAAKE Minilab II twin-screw mixer (Thermo Fisher Scientific, Waltham, MA, USA)) ($T = 185$ °C, $V_{\text{screw speed}} = 30$ rpm, $t_{\text{mixing mode}} = 5$ min).

Melt quality was examined using a microscope with a heated stage (Boetius microscope, VEB Kombinat Nadema, Ruhla, Germany, former GDR).

Macrofibers were formed by extruding the melt through a die. The fiber winding speed was 3 m/min, and the drawing ratio was 6.2.

To simulate the processing of secondary PP, the macrofibers were crushed (up to 3 mm), then loaded into an extruder together with PG, actively mixed at 185 °C ($t = 15$ min). The resulting melt was passed through a spinneret to create macrofiber. We classify the proposed conditions as model (laboratory) conditions, which should be taken into account during further scaling of the process, where it is possible to change the conditions for processing systems with PG, for example, temperature-time regimes, etc.

2.3. Characteristics of Polypropylene Fibers

X-ray diffraction (XRD) and IR spectroscopy were used to evaluate the structural features and chemical changes in PP fibers. XRD analysis of the macrofibers was performed on a TD-3700 diffractometer (Dandong Tongda Science & Technology, Dandong, China) (linear focus 1×10 mm, source operating mode 40 kV-30 mA, CuK α $\lambda = 1.542$ Å) and a linear semiconductor detector. Diffraction patterns were recorded at room temperature in transmission mode using the Bragg–Brentano scheme in continuous scanning mode θ - θ in the angular range of 5–35°, with a scanning step of 0.02° and an intensity accumulation time of 1 s per point.

The evolution of strength depending on the history of PP fibers was studied using an Instron 1122 tensile testing machine (Instron, Norwood, MA, USA). The strain rate of the macrofibers was 500 mm/min. For tensile testing, the macrofibers were cut into 30 mm long staple fibers. The initial distance between the grips was 10 mm. The macrofiber diameter was measured using an electronic micrometer IP65 (Mitutoyo Digimatic, Kanagawa, Japan).

All measurements were performed in triplicate, and the average value is indicated. At least three batches of each PP processing cycle were used for mechanical testing. Each batch contained at least 20 macrofiber samples.

3. Results and Discussion

3.1. Morphology of Melts

The virgin PP is colorless, and the granules are semitransparent. During PP processing, depending on the temperature and intensity of mechanical stress, the polymer may change color and acquire a yellowish tint [51]. Increasing the number of PP processing cycles intensifies the color change. This color change is caused by chemical reactions occurring at high temperatures and a decrease in the length of the macromolecules. Figure 2 shows the morphology of PP melts with propyl gallate.

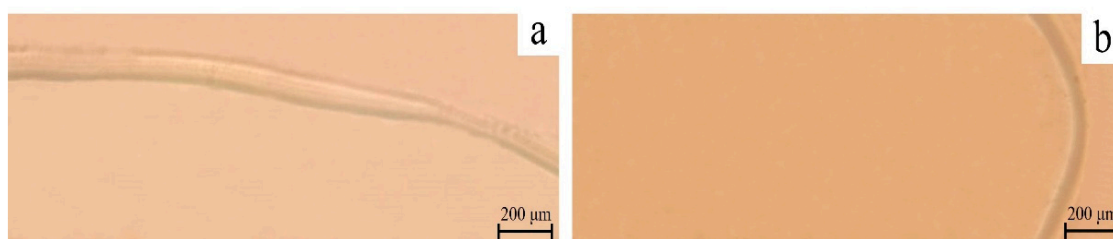


Figure 2. Morphology of PP melts with propyl gallate after 1 (a) and 5 (b) passes.

The photographs show that the melt's appearance changes only slightly. The melt with the antioxidant remains transparent after five passes. However, its color changes slightly, acquiring a slightly yellowish tint. It is important that the addition of the antioxidant to PP results in a uniform melt morphology without visible inhomogeneities or agglomerates.

Figure 3 shows photographs of the appearance of cooled PP melts with and without stabilizer additives.

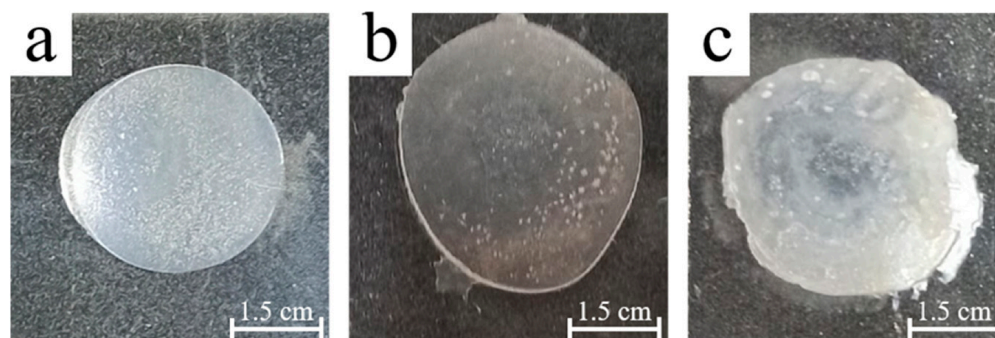


Figure 3. Melts of PP of the virgin (a) and with propyl gallate after 1 (b) and 5 (c) passes.

In the work [52], it is shown that after five recycling cycles, PP significantly changes its appearance. The presented photographs show that after numerous recycling cycles, the PP + PG system changes its appearance only slightly. Thus, the introduction of the antioxidant PG into melts helps preserve the color and appearance of the polymer that has undergone multiple recycling. Compared to melts with an antioxidant, the original polymer acquires a slight yellowness after the first recycling cycle. After five recycling cycles, PP yellowness intensifies, but the melt still retains transparency. When using propyl gallate, the melts differed radically in color from the control PP melts.

After five passes accompanied by exposure to high temperatures and mechanical deformation of the samples, melts with propyl gallate are characterized by minor color changes, in contrast to the original PP. When reprocessing PP melts without a stabilizer, a change in the molecular weight of the polymer is possible, as well as the formation of secondary degradation products, which are the cause of the color change of the polymer. Thus, for systems based on low-pressure polyethylene, stereoisomers of various oxidation products are formed, such as dihydroxy dimers, spirocimers, trimers and aldehydes [53]. Although the color change indicates changes in the polymer, it does not limit its processing. It is important to note that the propyl gallate additive is uniformly distributed, as evidenced by the homogeneous pattern of the melts. Consequently, the ability of the melt to spin fiber will not be complicated by the presence of residual additive particles and will be determined by the rheological properties of the system.

3.2. Rheological Properties of Melts

Rheological studies allow us to quickly determine the effect of antioxidant additives on the preservation of the viscous and elastic properties of polypropylene melts. Thermal degradation of polyolefins results in a sharp decrease in viscosity, depending on the number of passes. Figure 4 shows the flow curves for PP melts without antioxidants and with PG additives.

The presented flow curves can be divided into two types based on their course. For the first type of curves, two regions of the flow curve are observed. The first region corresponds to the Newtonian region, where the viscosity does not change with increasing shear rate (shear-thinning (pseudoplastic behavior)). The second region is the structural branch; here, at high shear rates, the melt structure is destroyed and the viscosity decreases. Virgin PP has a melt viscosity of approximately 10^4 Pa s. The first PP pass does not lead to a significant decrease in viscosity. Multiple processing of PP without an antioxidant leads to a decrease in viscosity to 10 Pa s (after 5 passes), which is probably due to thermooxidative destruction. The flow curves for PP melts after 5 passes are characterized only by the Newtonian region; the structural branch is absent from the flow curve (second type). With the introduction of PG into the system, the viscosity of the melts decreases slightly during the first processing cycle. After three cycles, the viscosity of the PP + PG system decreases

by only half an order of magnitude. Finally, after five passes, the viscosity of PP melts with antioxidant decreases to 2×10^3 Pa s. The flow curves for systems with PG remain virtually unchanged. With an increasing number of processing cycles, the origin of the structural branch shifts toward the region of higher shear rates. An increase in the Newtonian region of the flow curve may indicate degradation in the polymer. However, this degradation is less pronounced in melts with an antioxidant compared to PP without PG. It is important to note that all flow curves are characteristic of polyolefin melts and reflect the polymer's history and properties.

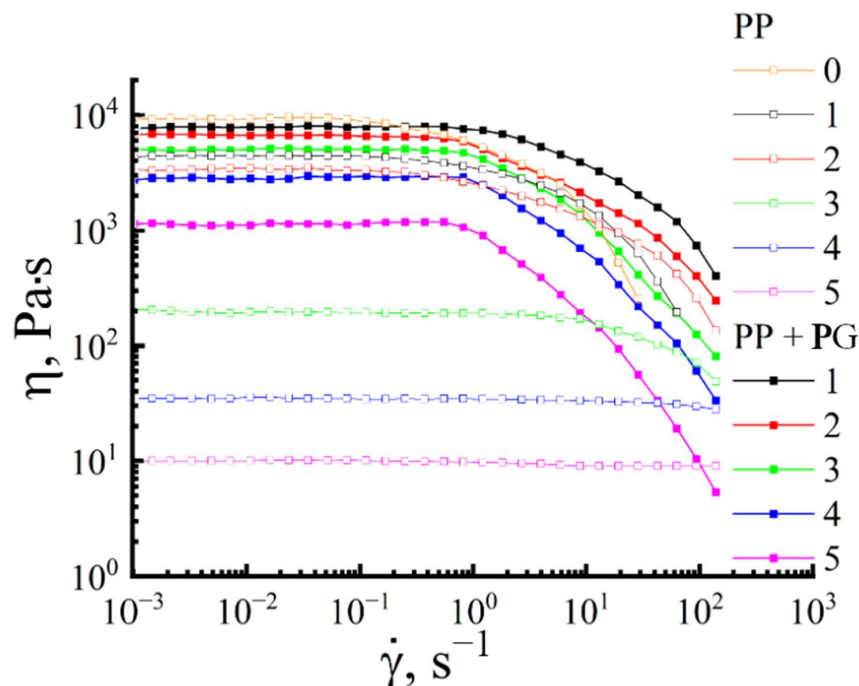


Figure 4. Flow curves of virgin PP melts with and without PG additives with different histories. The number of passes is indicated in the figure, $T = 185$ °C.

To more clearly illustrate the influence of PG on viscosity evolution, let us consider the dependence of melt viscosity on the number of processing cycles (Figure 5).

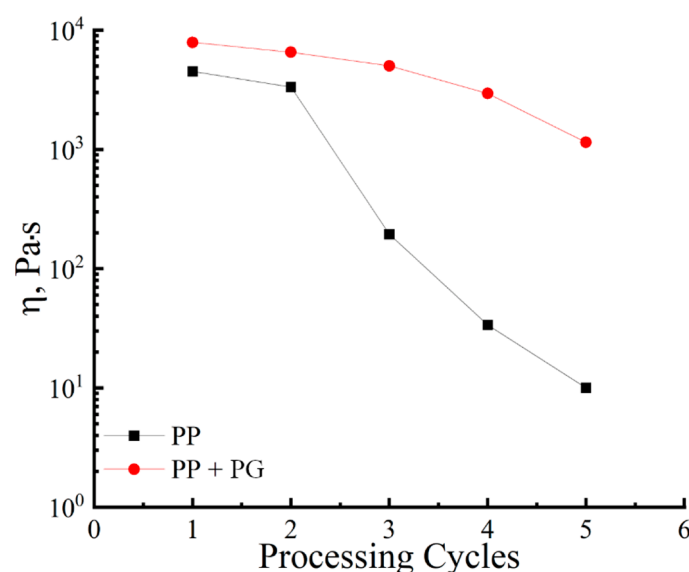


Figure 5. Dependence of melt viscosity on the number of processing cycles at a temperature of 185 °C.

The dependence of viscosity on the number of processing cycles shows that the first processing cycle does not reveal a significant decrease in viscosity. Already by the third processing cycle, a sharp decrease in viscosity is observed for PP melts, continuing until the last cycle. In the case of melts stabilized with PG, their viscosity decreases, but at a significantly slower rate. The decrease in the polymer molecular weight in PP melts causes a viscosity reduction of more than two orders of magnitude. After five passes, the difference between systems with PG and without antioxidant is also more than two orders of magnitude. Thus, the nature of both the flow curves and the viscosity dependence on the number of passes indicates the antiradical activity of the additive. PG provides a protective effect for PP melts and helps maintain viscosity values.

To quantitatively assess the effect of multiple processing on the viscoelastic properties of melts, dynamic studies of melts were carried out (Figure 6).

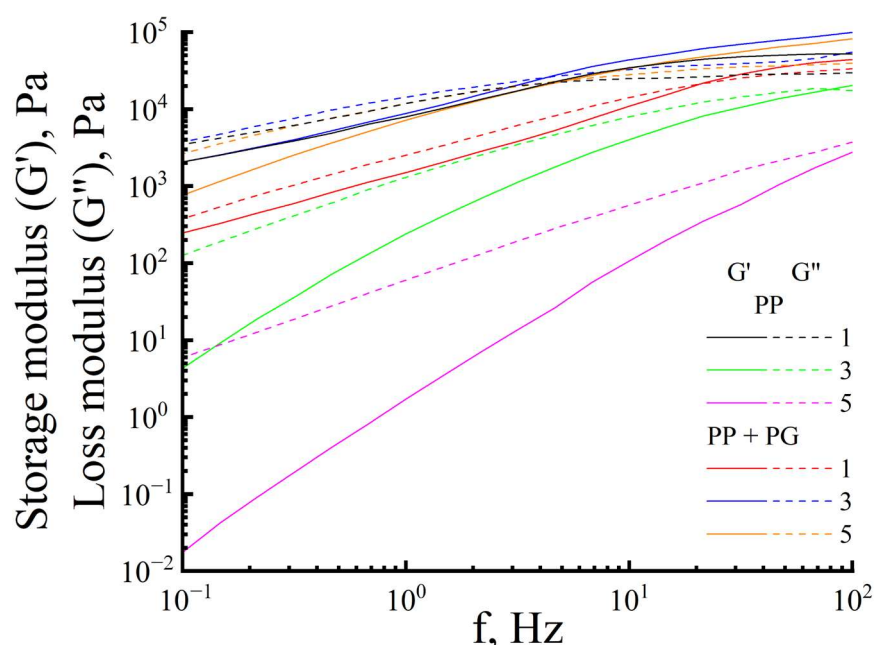


Figure 6. Dependences of the elastic modulus (G') and loss modulus (G'') on frequency for PP and PP + PG melts. The number of passes is indicated in the figure, $T = 185\text{ }^{\circ}\text{C}$.

The presented frequency dependences of the elastic moduli G' and loss moduli G'' have a characteristic shape for polymer melts. With an increase in the number of passes, the slope of the G' and G'' curves at low deformation frequencies increases. Moreover, the rate of change in the slope angle depends on the system composition and the number of passes. For PP melts without an antioxidant, after the fifth cycle, the difference in the rate of decrease in the slope of the curves leads to the fact that they do not intersect in the entire studied range of deformation frequencies. This may indicate an increase in the viscous component compared to the elastic one. At low deformation frequencies, $G' < G''$, with a further increase in the deformation frequency, a crossover point is observed at which $G' = G''$. Dynamic studies revealed a shift in the crossover point (the intersection of the frequency curves for the moduli G' and G'') to the region of higher frequencies for pure PP with an increase in the number of passes, which indicates a reduction in the relaxation time due to the destruction of polymer chains. In the presence of PG, a different picture is observed; the shift in the crossover point is less pronounced, which confirms the preservation of the elastic properties of the melt and the length of the macromolecules.

Figure 7 shows the dependence of the frequency at which the crossover point is observed on the number of processing cycles.

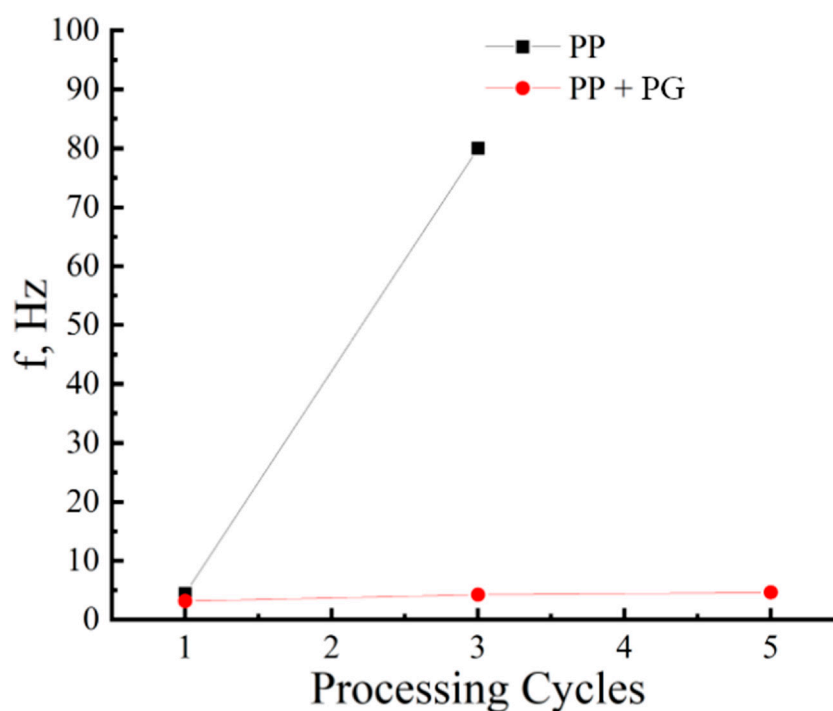


Figure 7. Dependence of the crossover point frequency on the number of melts passes.

The presented dependence shows a sharp shift in the frequency crossover point for PP melts from 10 to 80 Hz for three cycles. As the number of passes increases to five, the crossover point shifts beyond the measurement range, and in the presented range, the melt is characterized by viscous-dominant properties. For melts with an antioxidant, the crossover point position remains virtually unchanged for all cycles. Thus, the introduction of PG into the melt allows its elastic properties to be maintained during repeated processing.

Based on the rheological studies conducted, a process flow was developed for producing macrofibers from recycled polypropylene with the addition of propyl gallate. The overall process flow diagram is shown in Figure 8.

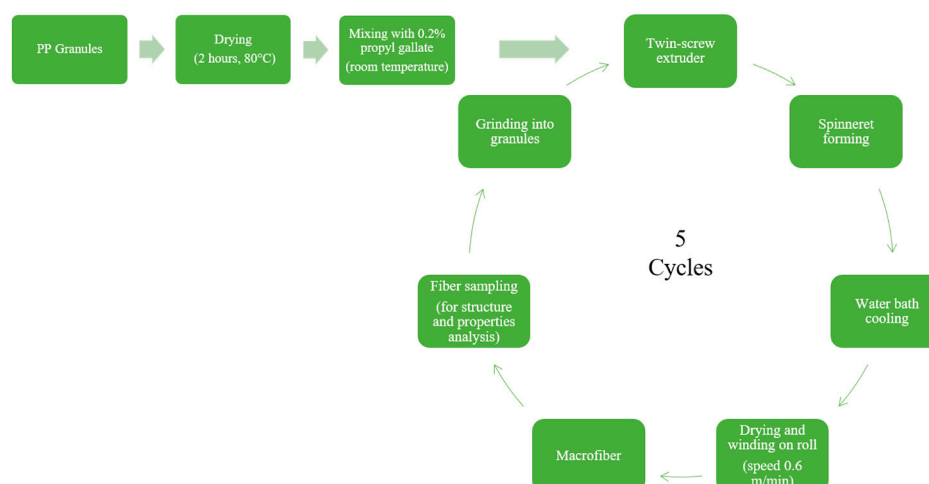


Figure 8. Schematic diagram of the process for producing macrofibers from polypropylene with the addition of propyl gallate.

According to the presented scheme, the use of primary PP together with PG to form molded products opens up the possibility of at least five cycles of polymer recycling.

We used the obtained rheological characteristics to select fiber formation modes. Macrofiber production was carried out on a laboratory bench equipped with a twin-screw extruder (Figure 9).

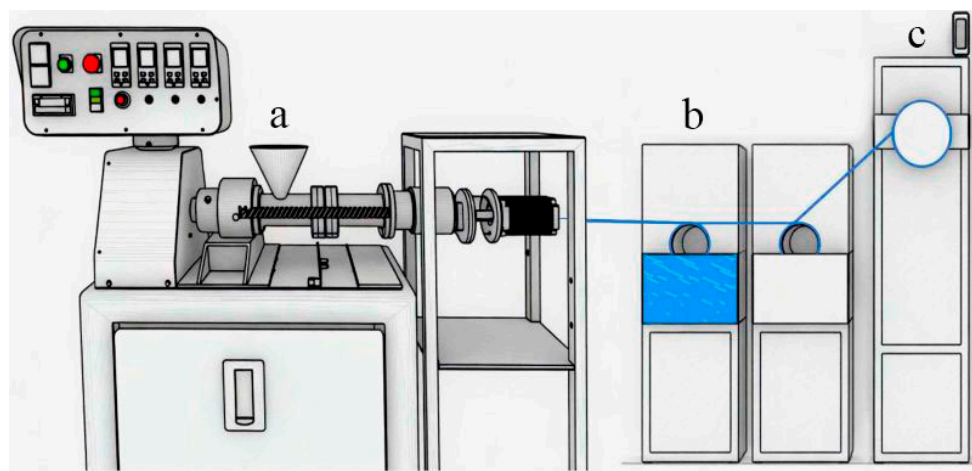


Figure 9. Macrofiber spinning setup: extruder (a), fiber cooling bath (b), winding shaft (c).

When spinning macrofibers, the extruder was loaded with PP and PG. Three heating zones ensured good mixing of the components. After exiting the die, the hot melt was directed into a cooling bath and then through guide rollers to the receiving device. Regardless of the melt history, the filaments formed reliably under the selected conditions. Examples of spun macrofibers are shown in Figure 10.

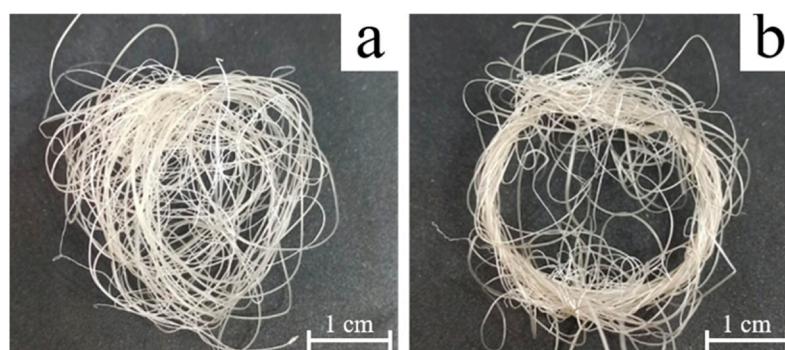


Figure 10. Photographs of spun PP + PG fibers (a,b) after 1 (a) and 5 (b) passes.

The resulting fibers have an average diameter of 140 micrometers. Their surface is smooth and even. The color of the resulting fibers reflects the morphological pattern of the melts described above; specifically, for PP + PG fibers, a yellowish tint appears after five processing cycles.

The morphology of the fibers in polarized light is shown in Figure 11.

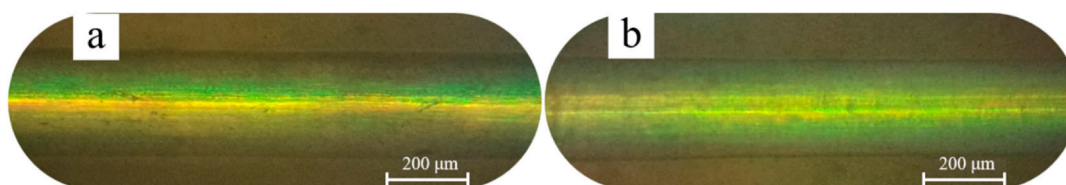


Figure 11. Morphology of fibers with the addition of propyl gallate in polarized light after 1 (a) and 5 (b) passes.

In crossed polaroid microscope images the highly ordered molecular structure of the polymer chains causes the fibers to exhibit birefringence. The observed optical pattern is maintained regardless of the number of recycling cycles.

3.3. Wettability of Melts

To assess the impact of repeated processing and the addition of propyl gallate on the surface properties of polymer melts, we measured the contact angle. Water and a 12% aqueous solution of sodium hydroxide (NaOH) were used as test liquids. Photographs of PP films with water droplets deposited on them are shown in Figure 12.

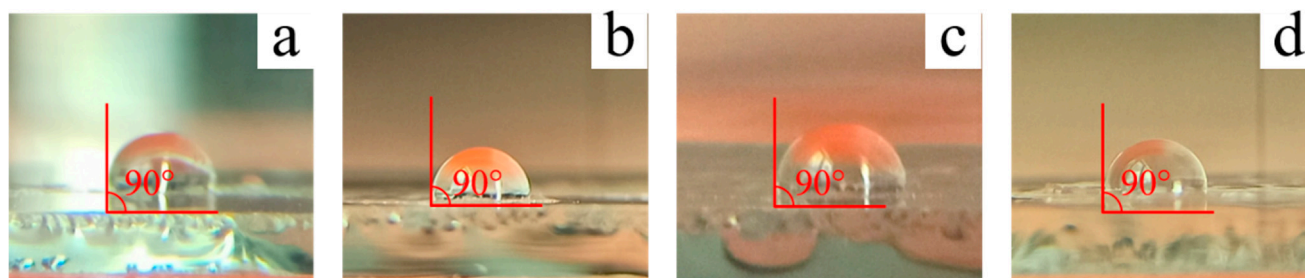


Figure 12. Water droplet profiles on PP melts without stabilizer (a,c) and with propyl gallate (b,d) after 1 (a,b) and 5 (c,d) passes.

When exposed to water, the contact angle for all tested samples was 90° regardless of the number of processing cycles (1 and 5) and the presence of propyl gallate in the melt. Since cement mortars are alkaline, aqueous alkali solutions were also applied to the PP film surface in addition to water. Photographs of droplets of a 12% aqueous NaOH solution on the PP film surface are shown in Figure 13.

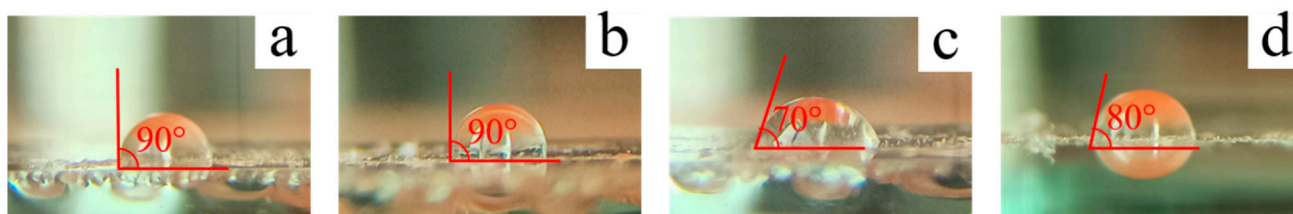


Figure 13. Profiles of droplets of 12% NaOH solution on PP melts without stabilizer (a,c) and with propyl gallate (b,d) after 1 (a,b) and 5 (c,d) passes.

The highest contact angle (90°) was found for PP without antioxidant and for the PP composite with propyl gallate after the first processing cycle. With an increase in the number of processing cycles to five for PP without antioxidant, the angle decreases by 20° (to 70°), while for the sample with propyl gallate, the angle decreases to 80° . Thus, PP fibers without an antioxidant will be better wetted by cement mortar compared to fibers from a stabilized melt.

3.4. IR Spectroscopy

PP is characterized by the following bands in the IR spectra: 2950, 2914, 2860, 2836, 1457, 1376, 1355, 1166, 997, 974, 898, 841, and 810 cm^{-1} . The interpretation of these bands is described in the work [54]. The spectra for PP macrofibers molded without the addition of an antioxidant and with PG are shown in Figure 14.

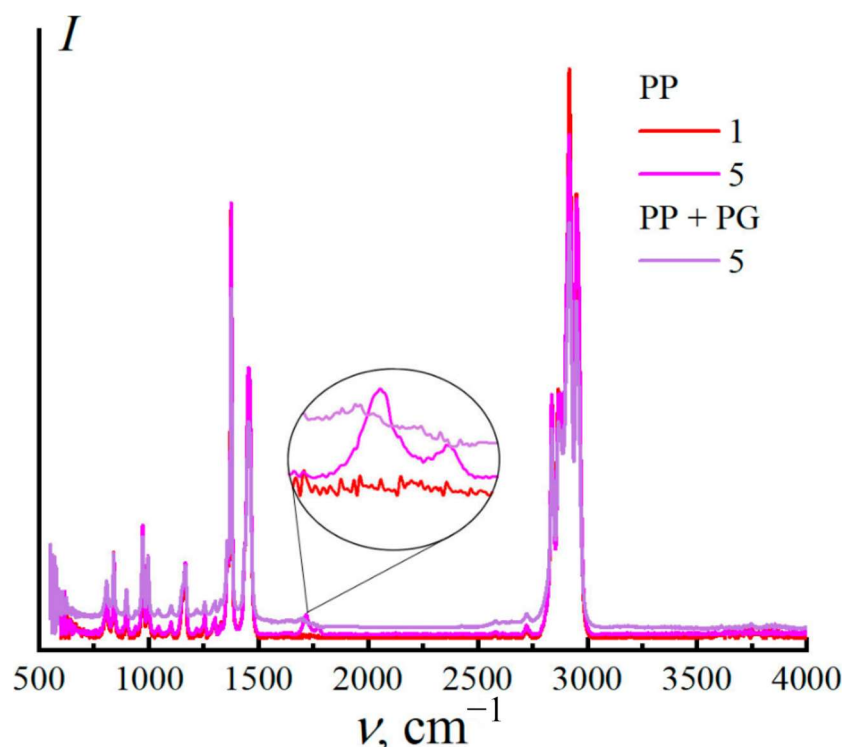


Figure 14. IR spectrum of PP macrofibers with different production histories (enlarged regions of the 1650–1800 cm^{-1} spectrum).

For recycled PP, the most interesting region is 1650–1800 cm^{-1} , which is shown in an enlarged form in Figure 11. With an increase in the number of recycling cycles, the intensity of the 1720 cm^{-1} band increases [55]. This is due to an increase in the number of carbonyl groups formed during oxidative degradation. For systems without PG, after five recycling cycles, the intensity of the 1720 cm^{-1} band is the highest compared to the melt after the first recycling cycle and the melt stabilized with an antioxidant after five recycling cycles.

The carbonyl index is often proposed for assessing PP degradation. A method described in detail, for example, in [56], is used to calculate the carbonyl index. Using this approach, it was found that the carbonyl index for the original polymer was several times lower than that for samples that had undergone five processing cycles without PG additives. With the addition of an antioxidant to PP, the carbonyl index values increased (0.07), but remained lower than those for systems without PG. It is important to note that the use of the carbonyl index is not always recommended for systems with stabilizing additives, as incorrect values may be obtained, which may also not reflect the deterioration in the mechanical properties of the polymer [57].

3.5. X-Ray Diffraction (XRD) Analysis

It is known that the structure of polypropylene can significantly differ based on the polymer's processing circumstances. Isotactic PP is recognized to occur in various crystalline forms, including α , β , γ , and δ (smectic). Among these forms, the α form is the most common and stable monoclinic crystal structure [58]. The characteristic reflections for the α -crystalline phase of polypropylene in the 2θ region are 14.3°, 17.1°, 18.7°, 21.8°, and 25.7°, corresponding to the planes (110), (040), (130), (131)/(041), and (060), respectively. Figure 15 shows the diffraction patterns for macrofibers spun from melts with PG additives.

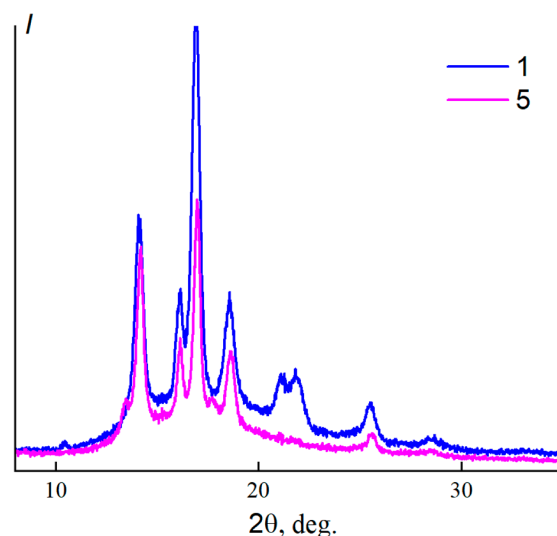


Figure 15. Diffraction patterns of PP + PG-based macrofibers (1 and 5 passes).

The angular positions of the main peaks remain unchanged for all samples. This indicates that no significant structural transformations occur during repeated processing—the polypropylene crystal lattice type remains unchanged. Differences are observed only in the peak intensities. The change in intensity is due to changes in the degree of crystallinity of the material. Therefore, it can be assumed that repeated processing does not lead to the formation of new crystalline modifications of polypropylene, but only affects the quantitative ratio of crystalline and amorphous phases. The crystallinity index of the fibers decreases with increasing number of recycling cycles. For fibers, the crystallinity index is 62% after the first cycle, while after five passes for the PP + PG system, it is 57%. The use of a stabilizer results in only an 8% decrease in crystallinity after several recycling cycles.

3.6. Mechanical Properties

The degradation of the polymer during repeated processing should be reflected in the mechanical properties of the spun fibers. Figure 16 shows the evolution of fiber strength depending on their history and the presence of an antioxidant.

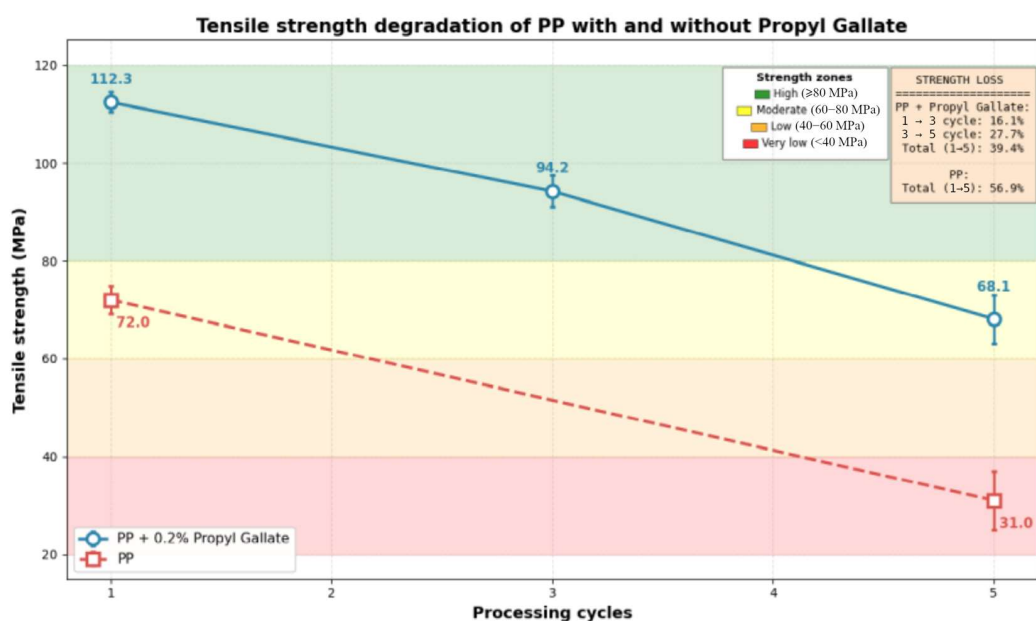


Figure 16. Dependence of fiber strength on their history and composition.

Increasing the number of PP passes leads to a decrease in fiber strength, regardless of the presence of an antioxidant. However, the use of PG allows for greater strength retention with increasing processing cycles. For PP macrofibers, after five processing cycles, the strength decreases from 72 MPa to 31 MPa, while for PP + PG systems, the decrease is from 112.3 MPa to 68.1 MPa. Since the mechanical properties of PP fibers depend on many factors, including the type of spinnerets used, the drawing ratio, post-processing of the spun fibers, and others, their mechanical properties can vary over a wide range, reaching several hundred MPa [59]. In the current study, a fiber spinning line was used that did not contain any additional processing steps for the spun PP fibers, and the drawing ratio was 6.2. Therefore, the obtained strength values for fibers with propyl gallate and without the addition of an antioxidant are underestimated. The observed strength of fibers with PG additives allows them to be considered for the formation of reinforced concrete structures, regardless of the number of passes.

Thus, the introduction of the antioxidant PG into PP helps maintain the viscous and elastic properties of melts, unlike systems without stabilizing additives. A similar effect is observed for PP with the addition of synthetic stabilizers, such as 0.75% Recyclobyk 4371 [60]. Since most popular stabilizers (Irganox and others) are synthetic, propyl gallate offers advantages because it can be obtained from natural raw materials. A more detailed comparative analysis of PG and Irganox will further reveal the advantages and disadvantages of the alternative natural stabilizer [61].

4. Conclusions

The study assessed the potential of the antioxidant propyl gallate as an effective stabilizer for polypropylene subjected to repeated thermomechanical processing. The addition of 0.2% propyl gallate to the PP formulation significantly slowed the characteristic oxidative degradation processes, which was comprehensively confirmed by the analytical methods used.

Rheological studies showed that the presence of propyl gallate promotes the preservation of the melt's viscoelastic properties. Unlike unstabilized polypropylene, a slight drop in viscosity was observed in the system with the additive after repeated polymer processing. A small shift in the crossover point on the frequency dependences of the elastic and loss moduli indicates greater preservation of the melt structure and probably the polymer's macromolecular length. Key evidence of the antioxidant effect was the significant suppression of carbonyl group accumulation in PP macromolecules, recorded by IR spectroscopy.

Visual analysis confirmed that propyl gallate slows the intense yellowing of the melt associated with degradation and promotes the formation of a uniform morphology without the formation of agglomerates of the introduced additive. It is important to note that despite the observed chemical changes, the rheological properties of the melt remained sufficient for a stable macrofiber formation process at all stages of cyclic processing.

The obtained results convincingly demonstrate that propyl gallate is a promising additive for stabilizing recycled polypropylene. Its use not only reduces the degree of polymer degradation during recycling but also preserves its processability, opening up opportunities for the creation of more sustainable and environmentally friendly polymer materials and products based on them.

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