


Influence of a biodegradable contaminant on the mechanical recycling of a low-density polyethylene sample

Vincenzo Titone¹  | Luigi Botta^{1,2} | Maria Chiara Mistretta^{1,2} | Francesco Paolo La Mantia^{1,2}

¹Department of Engineering, University of Palermo, Palermo, Italy

²INSTM, Consortium on Materials Science and Technology, Florence, Italy

Correspondence

Francesco Paolo La Mantia, Department of Engineering, University of Palermo, V. le delle Scienze, 90128 Palermo, Italy.
Email: francescopaolo.lamantia@unipa.it

Abstract

Mechanical recycling of oil-derived polymers is certainly our best option to reduce pollution, save raw materials, and protect ourselves and the environment from the adverse effects of waste disposal. However, the presence of contaminants, including other types of plastics, that are mixed in during the recycling collection process or during the mechanical waste sorting stage could adversely affect the quality of the recycled product, leading to the recycling of a poor-quality secondary material. In this work, the influence of a biodegradable contaminant on the mechanical recycling of a low-density polyethylene (LDPE) sample was investigated by rheological (shear and non-isothermal elongation) and mechanical analyses. The results showed that 2% of the contaminant is able to influence the rheological, shear, and isothermal elongation properties of recycled LDPE, while the results of the mechanical tests showed that after one extrusion cycle, the main tensile properties were not significantly affected by the presence of the contaminant, but after only two cycles of extrusions, some significant reduction in the final properties began to appear. In short, the presence of 2% of a biodegradable co-polyester in a LDPE matrix gives rise to a more pronounced decay of the rheological and mechanical properties, but, after two extrusion steps, both rheological and mechanical properties seem still useful for the production of film.

Highlights

- To what extent the contaminant affects the quality and usability of the recycled LDPE;
- The effects of contamination on the LDPE sample during the recycling process;
- Reduction in properties, compromising product quality;
- Compromised product quality after only two additional processing steps;
- Important implications for the recycling industry.

This is an open access article under the terms of the [Creative Commons Attribution](https://creativecommons.org/licenses/by/4.0/) License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

© 2023 The Authors. *Polymer Engineering & Science* published by Wiley Periodicals LLC on behalf of Society of Plastics Engineers.

KEYWORDS

biodegradable polymer, contaminant, poly(butylene adipate-co-terephthalate) (PBAT), polyethylene (LDPE), recycling

1 | INTRODUCTION

Mechanical recycling is one of the main technologies used in industry to process processing waste and post-consumer polymers,^{1–3} and it is a key process for reducing environmental impact and promoting sustainable management of plastic waste.⁴ Moreover, it is a fairly inexpensive, large-scale, solvent-free technology applicable to all the polymers.^{5–12} However, for the quality of the resulting products to be high, a sorting process must be carried out that provides clean recycling material by removing any foreign fractions such as biodegradable polymers. Certainly, recycling materials of different chemical nature raises some specific issues that need to be addressed,^{13–19} just as it is important to carefully consider the presence of contaminants during the collection of waste for recycling in order to take the necessary measures to mitigate undesirable effects.

Biodegradable polymers are designed to biodegrade over time when exposed to specific environmental conditions. Although the biodegradable polymers can be recycled like all the other conventional polymers, composting is an important alternative way for the end of life of these polymers. This means that biodegradable polymers must be separated from the other polymers. On the other side, the presence of biodegradable polymers in the stream of non-biodegradable polymers can cause some damage to the quality of the recycled polymers.^{20–22} In fact, many biodegradable polymers can degrade during processing, making it difficult to achieve good quality of the recycled material. In addition, they may have a different melting temperature than conventional polymers, which can complicate the recycling process.

Poly(butylene adipate-co-terephthalate) (PBAT) is a biodegradable polymer obtained from the polymerization of alkyl alcohols, terephthalic acid, and adipic acid. It is characterized by the presence of ester bonds in its structure, which gives the material considerable flexibility and mechanical strength.²³ In addition, it is a highly flexible material with good weather and UV resistance, making it suitable for various applications such as the production of composting bags, biodegradable food films and coatings, packaging, and environmentally sustainable disposable products.^{24–26}

Low-density polyethylene (LDPE), on the contrary, is, in the plastics industry and particularly in the packaging sector, the most widely used material due to its distinctive

characteristics, such as density and flexibility, and it also turns out to have the highest recycling rate.^{27–29} However, LDPE, being a widely used but non-degradable packaging material, poses a serious pollution threat to the environment. For sustainability purposes, the new directive requires that most products from municipal waste collection must be recycled (mechanically or chemically) into value-added products. Consequently, also based on the above, if not well separated, the presence of contaminant could adversely affect the quality of the recycled product, leading to the recycling of a poor-quality material and making the recycling process less efficient. In this regard, one of the potential contaminants could be PBAT itself, as previously reported, since it is a new degradable polymer widely used in packaging because it has properties very similar to LDPE.

The aim of this paper was to evaluate the influence of small amounts of a biodegradable contaminant, such as PBAT, during reprocessing on the rheological and mechanical properties of a low-density polyethylene for film blowing. In particular, recycling was simulated through successive extrusion steps. The co-polyester undergoes mainly hydrolytic chain scission during the extrusions giving rise to small chains with respect to the long macromolecules of the low-density polyethylene that, not only are incompatible, but also act as lubricant decreasing the viscosity of the polymer systems.

Processability and mechanical properties of the LDPE are worsened by the presence of this small amount of PBAT and the mechanical properties of the contaminated sample after two recycling steps are very similar to those measured on the recycled pure LDPE after five recycled steps. However, both rheological and mechanical properties seem still compatible with a film blowing operation and with the typical properties of film for packaging after two reprocessing steps.

2 | EXPERIMENTAL

2.1 | Materials

The low-density polyethylene (LDPE) used in this work was a film blowing grade Riblene[®] FC30 supplied by Versalis (Italy) having the following properties: melt flow index (M.F.I.) = 0.27 g/10 min (at 190°C, 2.16 kg loading), density = 0.922 g/cm³, and melting temperature \cong 113°C.³⁰

TABLE 1 Composition and recycling steps of investigated samples.

Sample Code	Composition of recycled samples
LDPE	ext-LDPE (100)
R	r-LDPE (100) extruded five times
REC	r-LDPE/PBAT (98:2) extruded two times

The contaminant used was a poly(butylene adipate-co-terephthalate) (PBAT) supplied by BASF (Germany) with the following properties: MFI = 2.7–4.9 g/10 min (at 190°C, 2.16 kg loading) and Mn = 24,400 g/mol.³¹ PBAT has been chosen because it is used in the same applications of LDPE, for example, for packaging and shows similar properties.

2.2 | Mechanical recycling of LDPE and LDPE/biodegradable contaminant

Mechanical recycling was simulated by multiple extrusions using a HAAKE PolyLab QC single screw extruder manufactured by Thermo Scientific (Karlsruhe, Germany).

Contaminant-free LDPE (R) was reprocessed up to 5 times, while LDPE (REC) in the presence of 2% of PBAT was reprocessed until 2 times. In both cases, the LDPE was extruded with a screw rotation speed of 60 rpm and with a temperature profile set at 150–160–170–180°C. Similarly, PBAT was extruded up to 2 times. As post-consumer LDPE is not pre-dried before recycling operations, PBAT was not dried before the process.

Table 1 shows the composition of recycled samples.

Figure 1 show example diagram of the following study.

2.3 | Characterization

Rheological characterization in shear flow was conducted using a model AR-G2 rotational rheometer (TA Instruments, New Castle, DE, USA) with parallel plate geometry. All measurements were performed at 180°C in an angular frequency range of 0.1 to 100 rad/s.

To evaluate the behavior of the presence of the pollutant and of the recycling steps on the filmability of the recycled material, non-isothermal elongational flow tests were performed using a capillary viscometer (Rheologic 1000, CEAST, Italy), equipped with a tensile module, operating at the same temperature as above. The melt strength of the melt filament is read directly as melt strength (MS), while breaking stretching ratio (BSR), the

ratio of drawing speed at break to extrusion speed at the die, was calculated, as reported in previous papers,^{32,33} as follows (Equation 1):

$$BSR = \frac{V_{roll}}{V_p} \cdot \frac{D_p^2}{D_c^2}, \quad (1)$$

where V_{roll} is the collecting speed, V_p is the capillary piston speed, D_p is the piston diameter, and D_c is the diameter of the capillary.

Tensile tests were conducted at room temperature using an Instron universal testing machine (Instron, mod. 3365, High Wycombe, UK) according to ASTM D638-14.³⁴ Rectangular specimens (length = 90 mm, width = 10 mm and thickness = 0.5 mm) were tested at deformation rate of 1 mm/min until 3% deformation. Subsequently, the speed of crosshead was increased to 100 mm/min until the specimen failed. The mean value of the mechanical tests with the corresponding standard deviation is the result of ten measurements. Elastic modulus, E ; tensile strength, TS ; and elongation at break, EB , were the outcomes of the tests.

All the obtained materials were hot pressed with a Carver laboratory hydraulic press (Carver, Wabash, IN, USA) at a temperature of 180°C with a mold pressure of 350 psi for about 3 min. The cooling time was 10 min.

3 | RESULTS AND DISCUSSION

Figure 2 shows the complex viscosity curves versus angular frequency. For simplicity, only the curves of the LDPE and the reprocessed R₃, R₅, REC, and REC₂. Furthermore, in the same figure, the flow curves of the PBAT and PBAT extruded two times in the same conditions are reported.

The viscosity of LDPE decreases with increasing the number of extrusions, and the reduction is greater for the sample processed 2 times in the presence of the contaminant. The PBAT undergoes also a dramatic decrease of the viscosity after two extrusion steps. The decrease of the viscosity of the LDPE is due to the thermomechanical stress undergone during the repetitive passages in the extruder. The decrease of the viscosity of PBAT can be due to the same reason but, more probably, to the hydrolytic degradation occurring at the high extrusion temperature in presence of humidity. Conversely, after two extrusions, the Newtonian viscosity of the contaminated LDPE sample shows a value similar to that of the pure LDPE extruded 5 times.

The remarkable decrease of the viscosity in presence of this small amount of PBAT can be interpreted

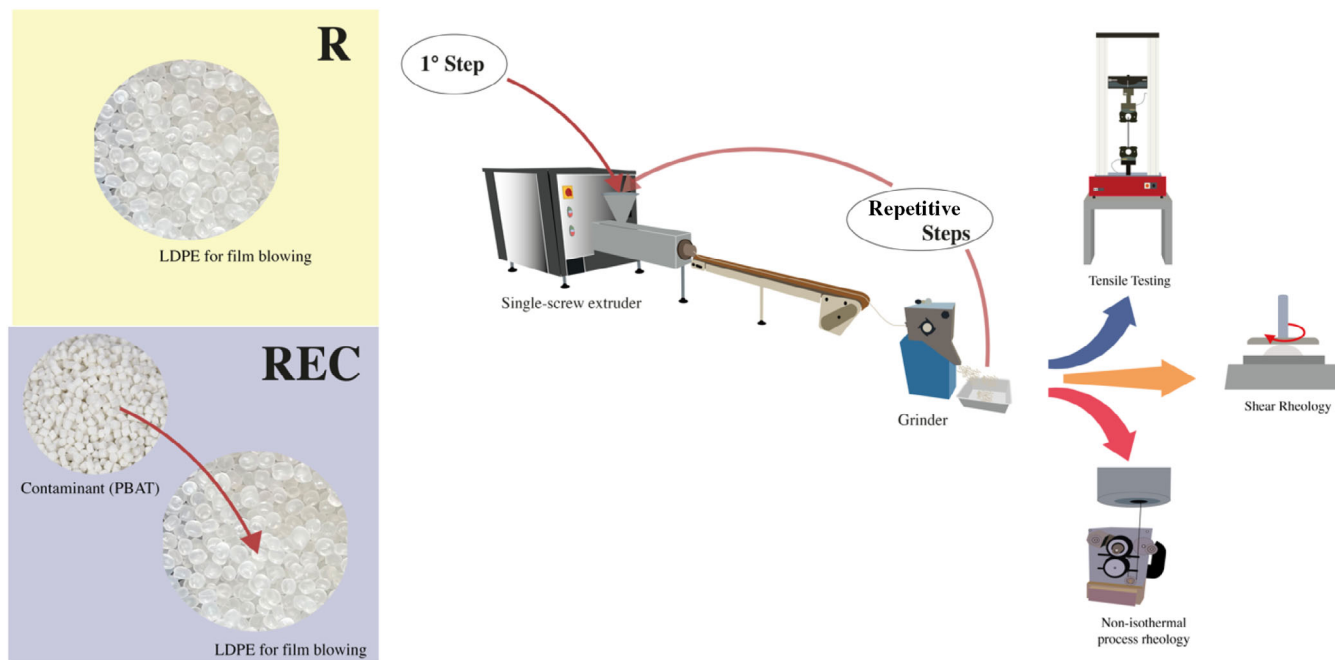


FIGURE 1 Example diagram of the processes used in this study.

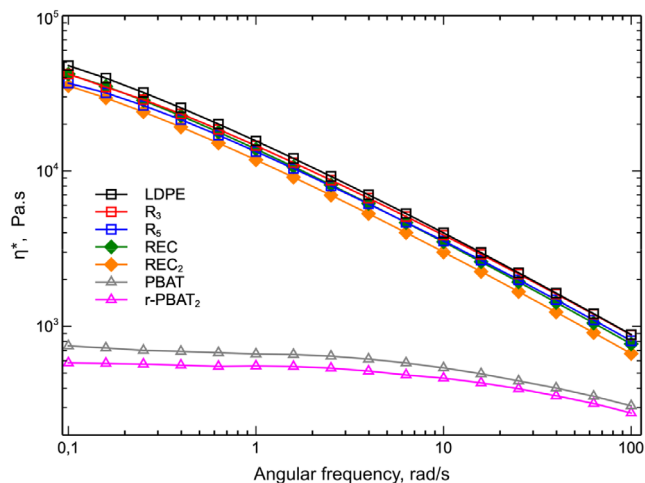


FIGURE 2 Flow curve of LDPE, PBAT, and the reprocessed R_3 , R_5 , REC, REC₂, and r-PBAT₂ samples.

considering the presence of small amounts of a very low viscosity component that, after two extrusion steps, shows a viscosity about 10 times lower than that of the matrix. This small amount can act not only as an incompatible component but also like a lubricant decreasing the viscosity of the blend.

Figure 3 show the storage modulus (G') versus angular frequency for LDPE and the reprocessed R_3 , R_5 , REC, and REC₂ samples.

It can be noted that the viscoelastic behavior is not affected by the addition of 2% by weight of the contaminant PBAT. In fact, the moduli values, in agreement with

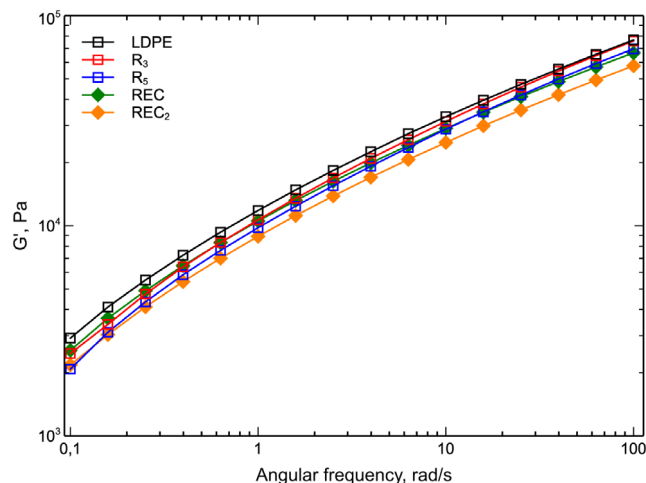


FIGURE 3 Storage modulus (G') versus angular frequency for LDPE and the reprocessed R_3 , R_5 , REC, REC₂.

TABLE 2 Initial slope of the G' curves of LDPE and of the reprocessed R_3 , R_5 , REC, and REC₂.

Property	LDPE	R_3	R_5	REC	REC ₂
Slope G'	0.601	0.640	0.668	0.611	0.671

the shear viscosity results, are slightly lower than LDPE, due to reprocessing, while the shape of the curves is almost unaffected by the presence of the contaminant. The initial slope of the G' frequency curves, see Table 2, slightly increasing from 0.601 for the LDPE to 0.640 for

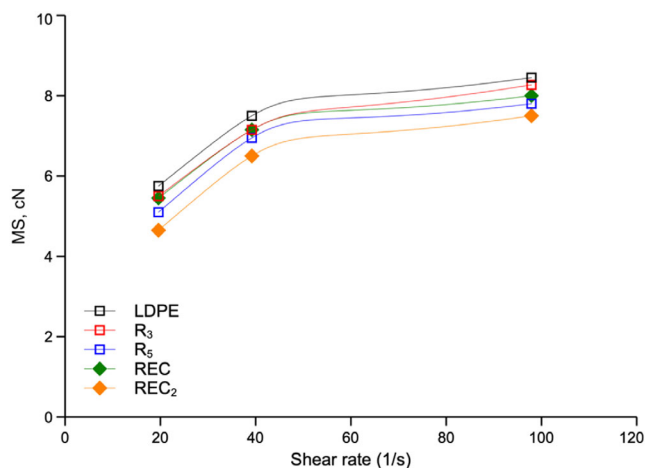


FIGURE 4 Melt strength (MS) versus apparent shear rate for LDPE and the reprocessed R₃, R₅, REC, and REC₂ samples.

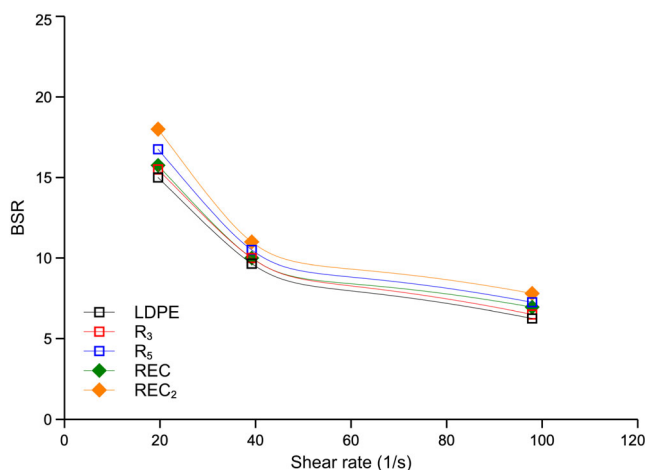


FIGURE 5 Breaking stretching ratio (BSR) versus apparent shear rate LDPE and the reprocessed R₃, R₅, REC, and REC₂ samples.

R₃ and 0.668 for R₅. The increase of the initial slope of the G' curves clearly put in evidence the increase of the viscous nature of the reprocessed samples and also in this case the behavior of R₅ and REC₂ are very similar.

As reported in the experimental section, in order to investigate the potential use of PBAT-contaminated r-LDPE for the same applications as neat LDPE, that is, film blowing process, a rheological characterization was carried out in non-isothermal elongational. Figures 4 and 5 show the measured MS and BSR of LDPE, and the reprocessed R₃, R₅, REC, and REC₂ samples, respectively.

Melt strength decreases as the number of reprocessing increases. In particular, the decrease in the sample reprocessed 2 times in the presence of contaminant (REC₂) is more pronounced than the variation in the sample reprocessed 5 times in the absence of contaminant (R₅), see

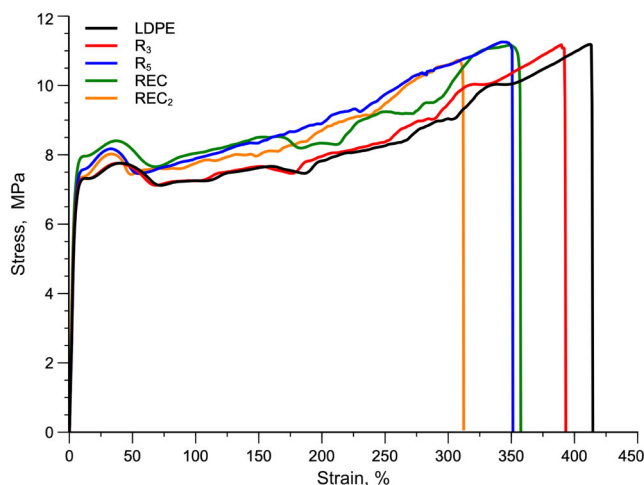


FIGURE 6 Typical stress-strain curve of LDPE and the reprocessed R₃, R₅, REC, and REC₂ samples.

TABLE 3 Mean tensile test results and the relative standard deviation of LDPE and the reprocessed R₃, R₅, REC, and REC₂ samples.

	E, MPa	TS, MPa	EB, %
LDPE	109 ± 8.9	11.1 ± 1.6	428 ± 19
R ₃	113 ± 7.2	11.0 ± 1.1	398 ± 15
R ₅	122 ± 12	11.0 ± 1.4	367 ± 16
REC	115 ± 9.1	11.0 ± 1.1	377 ± 15
REC ₂	125 ± 14	10.5 ± 1.3	341 ± 26

Figure 4. These results are in complete agreement with those of shear viscosity. Conversely, the increase in BSR of R₃, R₅, and REC is practically insignificant, while slightly more pronounced is that of sample REC₂, due to a more pronounced decrease in molecular weight resulting in a more deformable melt, see Figure 5.

Typical stress-strain curves and the main tensile test results are presented in Figures 6 and summarized in Table 3.

The average values of elastic modulus, tensile strength, and elongation at break of LDPE were 109 ± 8.9 MPa, 11.1 ± 1.6 MPa, and 428 ± 19%, respectively. As can be seen from the stress-strain curve, see Figure 6, the number of extrusion cycles has a clear influence mainly on elongation at break, while less relevance is observed on elastic modulus and tensile strength. Furthermore, in samples reprocessed in the presence of contaminant, this influence is more pronounced. More details, the elastic modulus increases by about 4%, 12%, 6%, and 14% for R₃, R₅, REC, and REC₂, respectively. This result, of the increase of the elastic modulus with the extrusion steps, was correlated⁷ with the increase of crystallinity due to the decrease of the molecular weight.

On the other hand, tensile strength remains almost unchanged, while a decrease in elongation at break is observed in all systems. In more detail, the elongation at break of LDPE was 428%, while R₃, R₅, REC, and REC₂ showed an elongation at break about 398%, 367%, 377%, and 341%, respectively. Of course, it is clear that the decrease, considering the number of reprocessing, are smaller for samples reprocessed in the absence of the contaminant than for those in the presence of the contaminant. In fact, this behavior, as already reported in the literature,²⁰ can be attributed to different phenomena such as immiscibility/incompatibility problems between the two components. The small particles of PBAT act as defects in the LDPE matrix giving rise to a more brittle polymer.

4 | CONCLUSION

In this paper, the influence of a biodegradable contaminant on the mechanical recycling of a low-density polyethylene sample has been investigated. Shear viscosity measurements revealed that viscosity decreases with the number of extrusion cycles. In addition, the presence of the contaminant leads to a more significant reduction of the viscosity than the recycling without pollutant. This is presumably due to the presence of small chains, originating from hydrolytic scission during extrusion, which, in addition to being incompatible, also act as a lubricant. Rheological tests in non-isothermal elongated flow have shown that the main rheological properties (and thus the performance during operations such as spinning and blowing) differ with the number of extrusion cycles. However, the rheological properties both in shear and in non-isothermal elongational flow of the contaminated LDPE sample after two extrusions are still compatible with a film blowing operation.

Finally, the mechanical test results allow us to observe that the main tensile properties, in the presence of the contaminant, begin to show some significant reduction only after two extrusion cycles. On the contrary, similar rheological and mechanical properties are shown by the uncontaminated LDPE after five recycling steps. The properties of the contaminated LDPE after two reprocessing steps, however, seem in the range of the properties typical of the film for packaging.

DATA AVAILABILITY STATEMENT

Data available on request from the authors.

ORCID

Vincenzo Titone  <https://orcid.org/0000-0001-5034-3675>

REFERENCES

- Gao F. Handbook of Plastics Recycling. F La Mantia(Ed). Rapra Technology, Shrewsbury UK, 2002. Pp 441, ISBN 1-85957-325-8. *Polym Int* 53, 233–233. 2004. doi:10.1002/pi.1323
- Schyns ZOG, Shaver MP. Mechanical recycling of packaging plastics: a review. *Macromol Rapid Commun*. 2021;42:2000415. doi:10.1002/marc.202000415
- Yin S, Tuladhar R, Shi F, Shanks RA, Combe M, Collister T. Mechanical reprocessing of polyolefin waste: a review. *Polym Eng Sci*. 2015;55:2899-2909. doi:10.1002/pen.24182
- Sadasivuni KK, Saha P, Adhikari J, Deshmukh K, Ahamed MB, Cabibihan J. Recent advances in mechanical properties of biopolymer composites: a review. *Polym Compos*. 2020;41:32-59. doi:10.1002/pc.25356
- Marrone M, La Mantia FP. Monopolymers Blends of virgin and recycled polypropylene. *Polym Recycl*. 1996;2:9-15.
- Dorigato A. Recycling of polymer blends. *Adv Ind Eng Polym Res*. 2021;4:53-69. doi:10.1016/j.aiepr.2021.02.005
- La Mantia FP, Mistretta MC, Titone V. An additive model to predict the rheological and mechanical properties of polypropylene Blends made by virgin and reprocessed components. *Dent Rec*. 2021;6:2. doi:10.3390/recycling6010002
- Wenguang MA, La Mantia FP. Recycling of post-consumer polyethylene greenhouse films: Monopolymer Blends of recycled and virgin polyethylene. *Polym Netw BLENDS*. 1995;5: 173-180.
- Valenza A, La Mantia FP. Recycling of polymer waste: part II—stress degraded polypropylene. *Polym Degrad Stab*. 1988; 20:63-73. doi:10.1016/0141-3910(88)90094-8
- Wenguang M, La Mantia FP. Processing and mechanical properties of recycled PVC and of homopolymer Blends with virgin PVC. *J Appl Polym Sci*. 1996;59:759-767. doi:10.1002/(SICI)1097-4628(19960131)59:53.0.CO;2-V
- Suhaimi NAS, Muhamad F, Abd Razak NA, Zeimaran E. Recycling of polyethylene terephthalate wastes: a review of technologies, routes, and applications. *Polym Eng Sci*. 2022;62: 2355-2375. doi:10.1002/pen.26017
- Mihut C, Captain DK, Gadala-Maria F, Amiridis MD. Review: recycling of nylon from carpet waste. *Polym Eng Sci*. 2001;41: 1457-1470. doi:10.1002/pen.10845
- La Mantia FP, Curto D. Recycling of degraded polyethylene: blends with nylon 6. *Polym Degrad Stab*. 1992;36:131-135. doi: 10.1016/0141-3910(92)90149-Y
- Khait K. Advanced recycling Technology for Unsorted Plastic Waste. *J Vinyl Addit Technol*. 1996;2:345-348. doi:10.1002/vnl.10152
- Titone V, Gulino EF, La Mantia FP. Recycling of heterogeneous mixed waste polymers through reactive mixing. *Polymers*. 2023;15:1367. doi:10.3390/polym15061367
- La Mantia FP, Gardette JL. Improvement of the mechanical properties of photo-oxidized films after recycling. *Polym Degrad Stab*. 2002;75:1-7. doi:10.1016/S0141-3910(01)00199-9
- Kazemi Y, Ramezani Kakroodi A, Rodrigue D. Compatibilization efficiency in post-consumer recycled polyethylene/polypropylene Blends: effect of contamination. *Polym Eng Sci*. 2015;55:2368-2376. doi:10.1002/pen.24125
- Incarnato L, Scarfato P, Acierno D. Rheological and mechanical properties of recycled polypropylene. *Polym Eng Sci*. 1999; 39:749-755. doi:10.1002/pen.11463

19. Nir MM, Ram A, Miltz J. Performance of reprocessed multi-layer LDPE/Nylon-6 film. *Polym Eng Sci.* 1995;35:1878-1883. doi:[10.1002/pen.760352307](https://doi.org/10.1002/pen.760352307)
20. La Mantia FP, Botta L, Morreale M, Scaffaro R. Effect of small amounts of poly(lactic acid) on the recycling of poly(ethylene terephthalate) bottles. *Polym Degrad Stab.* 2011;97:S0141391011003582. doi:[10.1016/j.polymdegradstab.2011.10.017](https://doi.org/10.1016/j.polymdegradstab.2011.10.017)
21. Samper M, Bertomeu D, Arrieta M, Ferri J, López-Martínez J. Interference of biodegradable plastics in the polypropylene recycling process. *Materials.* 2018;1886:11. doi:[10.3390/ma11101886](https://doi.org/10.3390/ma11101886)
22. Aldas M, Pavon C, De La Rosa-Ramírez H, et al. The impact of biodegradable plastics in the properties of recycled polyethylene terephthalate. *J Polym Environ.* 2021;29:2686-2700. doi:[10.1007/s10924-021-02073-x](https://doi.org/10.1007/s10924-021-02073-x)
23. Okada M. Chemical syntheses of biodegradable polymers. *Prog Polym Sci.* 2002;27:87-133. doi:[10.1016/S0079-6700\(01\)00039-9](https://doi.org/10.1016/S0079-6700(01)00039-9)
24. Ferreira FV, Cividanes LS, Gouveia RF, Lona LMF. An overview on properties and applications of poly(butylene adipate-Co-terephthalate)-PBAT based composites. *Polym Eng Sci.* 2019;59:E7-E15. doi:[10.1002/pen.24770](https://doi.org/10.1002/pen.24770)
25. Kilic NT, Can BN, Kodal M, Ozkoc G. The potential use of epoxy-POSS as a reactive hybrid compatibilizers for PLA/PBAT Blends: “effect of PBAT molecular weight and POSS type”. *Polym Eng Sci.* 2020;60:398-413. doi:[10.1002/pen.25295](https://doi.org/10.1002/pen.25295)
26. Jian J, Xiangbin Z, Xianbo H. An overview on synthesis, properties and applications of poly(butylene-adipate-Co-terephthalate)-PBAT. *Adv Ind Eng Polym Res.* 2020;3:19-26. doi:[10.1016/j.aiepr.2020.01.001](https://doi.org/10.1016/j.aiepr.2020.01.001)
27. Ronca S. Polyethylene. *Brydson's Plastics Materials.* Elsevier; 2017:247-278 ISBN 978-0-323-35824-8.
28. Piergiovanni L, Limbo S. Plastic packaging materials. *Food Packaging Materials.* SpringerBriefs in Molecular Science; Springer International Publishing; 2016:33-49 ISBN 978-3-319-24730-4.
29. Teck Kim Y, Min B, Won Kim K. General characteristics of packaging materials for food system. *Innovations in Food Packaging.* Elsevier; 2014:13-35 ISBN 978-0-12-394601-0.
30. Technical Data Sheet Riblene FC 30. Accessed June 15, 2023. <https://www.granulat.com.pl/medi/pdf/Riblene%20fc%2030.pdf>
31. Mistretta MC, La Mantia FP, Titone V, Botta L, Pedferri M, Morreale M. Effect of ultraviolet and moisture action on biodegradable polymers and their blend. *J Appl Biomater Funct Mater.* 2020;18:228080002092665. doi:[10.1177/2280800020926653](https://doi.org/10.1177/2280800020926653)
32. Titone V, Mistretta MC, Botta L, La Mantia FP. Toward the decarbonization of plastic: Monopolymer Blend of virgin and recycled bio-based, Biodegradable polymer. *Polymers.* 2022;14:5362. doi:[10.3390/polym14245362](https://doi.org/10.3390/polym14245362)
33. Gigante V, Aliotta L, Coltelli MB, et al. Fracture behavior and mechanical, thermal, and rheological properties of biodegradable films extruded by flat die and Calender. *J Polym Sci.* 2020; 58:3264-3282. doi:[10.1002/pol.20200555](https://doi.org/10.1002/pol.20200555)
34. International, A. *Standard Test Method for Tensile Properties of Plastics.* ASTM International; 2014.

How to cite this article: Titone V, Botta L, Mistretta MC, La Mantia FP. Influence of a biodegradable contaminant on the mechanical recycling of a low-density polyethylene sample. *Polym Eng Sci.* 2024;64(2):845-851. doi:[10.1002/pen.26588](https://doi.org/10.1002/pen.26588)