Available online at www.sciencedirect.com

ScienceDirect

Procedia CIRP 122 (2024) 97–102

www.elsevier.com/locate/procedia

31st CIRP Conference on Life Cycle Engineering (LCE 2024)

Influence of the material history on the properties of recycled glass fiber reinforced polypropylene - impact of screw speed during injection molding

Lisa Dahrmann[a,1], Robert Kupfer[a], Maik Gude[a]

[a] *TUD Dresden University of Technology, Institute of Lightweight Engineering and Polymer Technology, Dresden, Germany*

[1] Corresponding author. *E-mail address: lisa.dahrmann@tu-dresden.de*

Abstract

The changing properties of thermoplastic injection molding compounds during mechanical recycling complicate their use in structural lightweight engineering applications. Above that, an impact of the often unknown processing history on the recycled materials can be assumed, since it influences the material structure and degradation. In the present work it is investigated, how the shear load in plastification affects the properties of recycled glass fiber reinforced polypropylene during multiple processing cycles. Based on a design of experiments five processing cycles with different screw speeds are performed resulting in 47 different processing variants (screw speed, processing cycle). The samples undergo mechanical (tensile, impact), rheological (Melt Volume Rate) and morphological (fiber length distribution) tests. It was determined that the influence of the processing cycle is crucial for material degradation and properties. Also the screw speed setting directly affects the processing behaviour since it influences the melt temperature and viscosity. However, for the chosen process parameters, no clear influence of the screw speed on the resulting mechanical, rheological and morphological properties is found. The study suggests that knowledge of the screw speed setting is not essential for circular economy scenarios, as long it does not exceed the common processing recommendations.

Keywords: circular economy; mechanical recycling; material degradation; injection molding; reprocessing; processing settings; glass fiber; polypropylene

---

1. Introduction

Approaches to resource saving and circular economy are becoming increasingly important in lightweight engineering [1]. Due to their low density and high strength lightweight materials like fiber-reinforced plastics can reduce the required material consumption [2]. This weight reduction can result in lower energy/fuel usage and, as a result, lower emissions during the use phase. This is particularly relevant in automotive applications. [3] Futhermore the recyclability of thermoplastic composites can help in saving resources and increasing circularity [4]. Compared to virgin materials, mechanical recycling allows a significant reduction of the material-related environmental impact of a product [5]. Especially short and long fiber reinforced thermoplastics can replace a broad range of construction materials in various technical applications and enable a circular design of lightweight structures [6]. Many technical lightweight structures are produced by injection molding using these materials because it allows for near-net-shape manufacturing [7]. In injection molding, secondary materials can be reprocessed easily [8]. Production waste including sprues, runners, or reject parts are shredded and reused in the injection molding process as regrind or regranulate [9]. As multiple processing can result in fiber damage and changes in the polymer structure, there is still uncertainty regarding the properties of recycled fiber reinforced thermoplastics [10]. Especially the influence of manufacturing settings during the multiple processing on the properties is rarely investigated. This uncertainty results in a limited use of recycled thermoplastic composites for highly stressed technical lightweight structures [11].

Various scientific investigations have been conducted on the impact of either recycling or process settings on the microstructure and the properties of thermoplastic materials. For example, González-González et al. investigated the influence of repeated extrusion processing on the polymer structure of non-reinforced polypropylene (PP). The results of their study indicate thermo-mechanical changes in the polymer structure. Rising numbers of extrusion cycles and higher processing temperatures lead to lower molecular weights and thus a higher flowability. [12]

10.1016/j.procir.2024.02.006

Colucci et al. studied the impact of mechanical recycling on short glass fiber reinforced polypropylene (PP-GF). The results show a decrease in stiffness and strength due to the reprocessing, which was primarily attributed to fiber shortening during recycling. [13] The processing-induced fiber shortening of PP-GF is confirmed by Gupta et al. using a single-screw extruder. According to their findings, the interactions between fiber, polymer and machine lead to bending of the fibers, resulting in fiber breaking. [14]
The available studies suggest that degradation may lead to a partial reduction in mechanical properties and changes in rheological and morphological properties. However, they also indicate the potential for mechanical recycling and repeated use of engineering plastics to enhance the sustainability of the life cycle in technical applications.

The impact of injection molding settings on the mechanical properties of varying mixtures of virgin and recycled polypropylene is demonstrated by Gu et al. They show that different processing settings have an effect on the resulting mechanical properties, depending on the amount of recyclate. [15]

Brast investigated the influence of processing parameters on long glass fiber reinforced polypropylene and found, that besides processing temperature, back pressure, and screw speed setting have a strong impact on fiber damage. [16]

Despite the solid knowledge on the individual effects, less is known about the influence of the injection molding settings on the properties of multiple recycled fiber reinforced materials.

In this study the effect of screw speed variation during multiple injection molding processing on the material properties of PP-GF is investigated for five cycles. In particular, it shall be clarified whether sequence and repetition effects are measurable. This knowledge could help answering the question, if processing induced material degradation accumulates in a way, that it is relevant for following life cycles. The longer these materials can be used in the value chain depends on the accurate assessment of their resulting material properties and the extent of material degradation.

2. Experimental

2.1. Material

The short glass fiber reinforced polypropylene homopolymer ALTECH PP-H A 2030/159 GF30 CP manufactured by MOCOM was used for the investigations. This material has been recycled in a closed loop without adding any virgin material or additives. It has a short glass fiber volume content of 30 %.

2.2. Methods

The material underwent processing in a standard injection molding machine (ARBURG, Allrounder 370H 290) with a screw diameter of 35 mm. The temperature profile in the injection molding process (220 °C-240 °C), the injection speed (16 cm^3/s) and the other setting variables were kept constant for all test series.

The PP-GF30 is processed five times to analyze the influence of multiple processing. To assess the impact of the machine settings, the screw speed was set to 5 m/min (low) or 18 m/min (high). A custom partial factorial design was built for the experiments (see Appendix A). The design of experiments (DOE) represents possible combinations of screw speed across multiple processing. The resulting 47 unique processing histories were designed to reveal potential sequence or repetition effects caused by the process setting and their impact on the material properties. The geometry of the shoulder bar specimens is defined by ISO 3167:2014-11. After every processing the material specimen underwent mechanical (tensile and impact tests), rheological (Melt Volume Rate (MVR)), and morphological (fiber length distribution) tests. According to the scope of testing the necessary amount of samples was taken after each injection molding. The remaining specimens were milled for the next processing cycle. For this purpose a toothed roller mill (WITTMANN, S-MAX 2) was used. The speed was 250 1/min. The regrind was reprocessed. The schematic diagram in figure 1 represents the workflow.

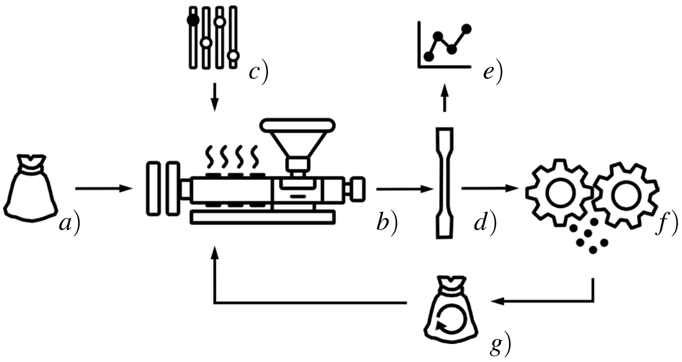

Fig. 1. Flow chart of the experimental test series, a) virgin material, b) injection molding, c) varying processing settings, d) samples, e) material properties, f) milling, g) recyclate

2.2.1. Fiber length distribution

The fiber length distribution was measured using the FASEP 3E Eco fiber scanner using the FASEP Analysis Software based on the Image Pro Plus 7.0 image processing program. It enables determination of the fiber length distribution and average fiber length. To analyze the fibers, they are separated from the matrix. The specimens underwent a complete thermoplastic matrix burn as it was subjected to incineration in a muffle furnace at 625 °C for four hours. The heating and cooling phases required an additional hour each, and the specimens were kept in the furnace for a total of six hours. The thermoplastic matrix burned completely. An average fiber volume content of 28 % was determined. The isolated fibers are then diluted with distilled water in a series of dilutions to approximately 0.025-0.035 g/l. One sample of each series was examined. Since the present study focuses on short fiber reinforced polypropylene, the number-averaged fiber length is determined to obtain an accurate picture of the fiber length distribution.

2.2.2. Melt Volume Rate

To evaluate the changes in viscosity, the Melt Volume Rate was determined after each processing cycle. The samples were prepared by using a manual shredder (Holi-Maker, HoliShred). The Melt Volume Rate was measured with a melt index tester (Göttfert, MP-B) according to the ISO 1133 and ISO 19069 standards. One MVR-test was performed per series.

2.2.3. Tensile tests

Six specimens of each test series underwent quasi-static tensile testing according to ISO 527-2. Stiffness, strength at break and elongation at break were analyzed. The Allround-Line testing machine Z250 (ZwickRoell) with a 10 kN load cell was used for the tests. The test speed was 5 mm/min. A contact extensiometer was used to record the elongation. The tests were performed under standard testing conditions at 23 °C and 50 % rel. humidity.

2.2.4. Impact tests

During production, retention samples were produced. These were used for orienting Charpy impact testing to provide a qualitative indication of material behavior. The test procedure followed ISO 179-1/fU. Deviating from the standard, however, only three test specimens could be tested. Only two were available for the single setting series in the first processing cycle. According to the standard, the specimens were cut from the shoulder bar tensile test specimens. The ZwickRoell Charpy impact testing machine (Zwick 5113) was equipped with a 15 J pendulum. The testing conditions were equivalent to the tensile tests.

2.2.5. Statistical evaluation

The statistical analyses were performed using JMP software to demonstrate the validity of the observations. To analyze the data, a least squares model was fitted. The DoE's variable factors are the number of processing cycles and the screw speed, both treated as continuous variables. The target variables to be maximized are Tensile stress at break, Young's Modulus and Number-averaged fiber length, while Elongation at break and Melt Volume Rate are to be minimized.

Using the JMP model, the statistically significant impacts of investigated factors on the target variables can be identified. For the evaluation of the created model, the probability value (P-Value) was considered to prove or refute the null hypothesis. The null hypothesis suggests that there is no significant relationship or effect between the variables being studied. A P-Value below 0.01 ($\alpha = 0.01$) indicates a correlation. The LogWorth is the negative logarithm (base 10) of the P-Value. It provides a more intuitive representation of the evidence against the null hypothesis. Once the value of 2 is reached, the null hypothesis is rejected.

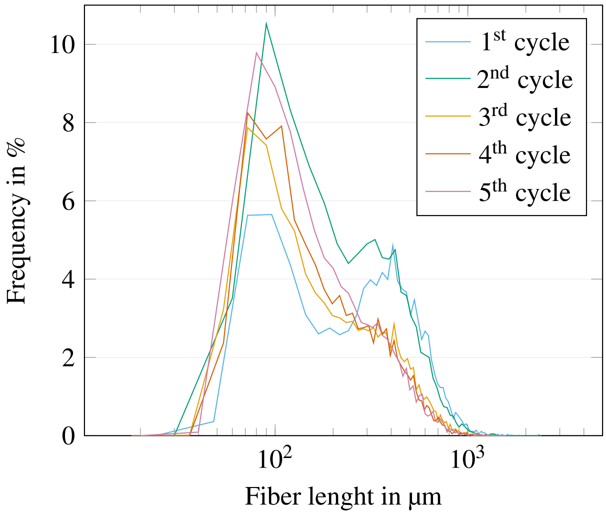

Fig. 2. Fiber length distribution of the high single setting series over five processing cycles

3. Results and discussion

The evaluation is categorized into single setting and mixed setting test series. In the single setting series, the process settings remain constant for all cycles (low or high). In the mixed setting series, the screw speed settings vary depending on the test design (low and high). An example of the test series can be found in Appendix A.

3.1. Morphological properties

Figure 2 exemplary shows the fiber length distributions for the five processing cycles of the single setting series with high screw speed. The content of longer fibers (300-500 µm) decreases rapidly in the first cycles. In contrast the fraction of short fibers (80-100 µm) rises with each processing cycle. The reduction in the frequency of longer fibers and the accumulation of shorter fibers during reprocessing can be seen.

Figure 3 compares the number-averaged fiber lengths of the single setting low and high series. The mean fiber lengths decrease about 45 % during the recycling cycles. The low series show slightly higher mean values in the number-averaged fiber lengths. The results are covered by general experience and physical models, which attribute the shortening of fibers and polymer chains to the shear stresses during injection molding. In consequence, higher screw speeds and multiple processing contribute lower average fiber lengths.

3.2. Rheological properties

The Melt Volume Rate was determined for both single setting series (low, high) and is shown in figure 4. A strong increase in MVR was observed over the multiple processing cycles from approximately 3 cm^3/10 min to 6 cm^3/10 min. The resulting increase in MVR is consistent with the literature and

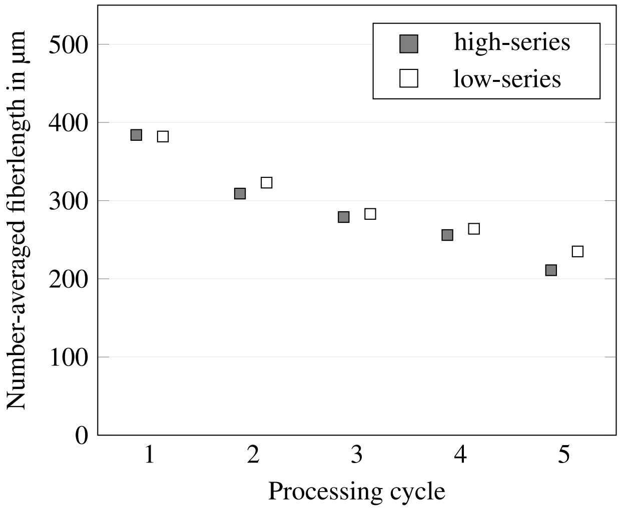

Fig. 3. Number-averaged fiber length of the single setting series (low, high) over the five cycles

also plausible given the decreasing average fiber length. [17] The MVR of the low series tends to be slightly lower than the high series. The only exception can be found in the third cycle, which could be an outlier e.g., caused by air trapped in the melt. However, the general increase in MVR with the cycle numbers is plausible before the background of the measured fiber length distributions (figure 3) and the known effect of molecular chain shortening.

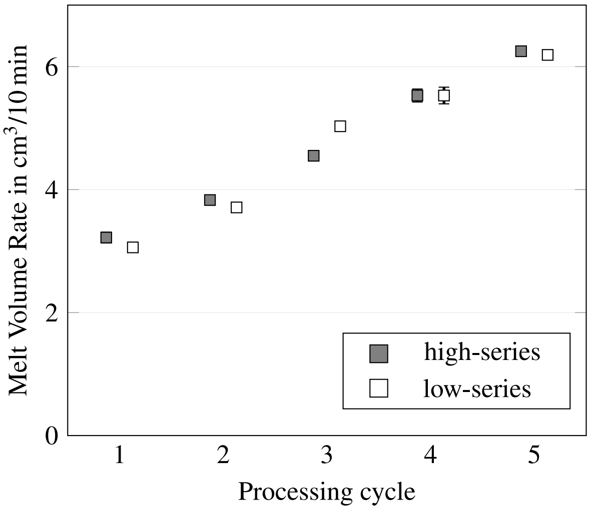

Fig. 4. Melt Volume Rate for the five processing cycles for the single setting series (low, high)

3.2.1. Processing behavior

Figure 5 shows the injection pressures of the low and high series for the five processing cycles. The values indicate the pressure which is necessary to completely fill the mold at the given injection speed. Lower injection pressures are measured at higher screw speeds across all cycles. The difference between the injection pressures of two following cycles decreases with rising cycle number.

Two physical effects are responsible here. First, higher screw speeds lead to increasing melt temperatures due to dissipation effects [18]. Since the polymer viscosity decreases with rising temperature, lower injection pressures are required to fill the mold in the high series. Second, high screw speeds are accompanied by strong shear stresses during plasticizing which contributes to fiber degradation [16]. As the results of the fiber length distribution and the according MVR indicate, the viscosity decreases with the fiber length, again reducing the required injection pressure in the high series. As seen in Fig. 2, the fiber length decreases most in the first two cycles and less in the latter. This is in line with the sinking differences between the injection pressures at higher cycle numbers.

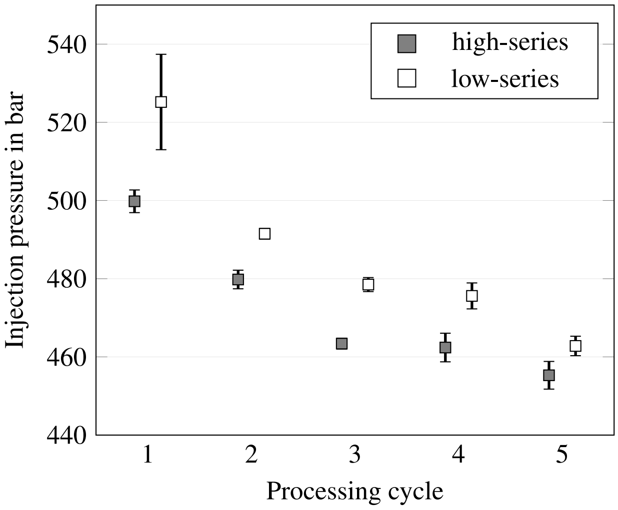

Fig. 5. Injection pressure values for the five processing cycles for the single setting series (low, high)

3.3. Mechanical properties

3.3.1. Tensile test

Figure 6 shows the Young's Modulus for the single setting series at all processing cycles. The box-plot diagram indicates the arithemetic mean, the 95 % confidence interval as well as the maximum and minimum values. The Young's Modulus decreases by approximately 16 % during the five reprocessing steps. No significant difference in the Young's Modulus can be observed between the series in each cycle, except of the fifth. Here a systematic error during the last measurement could be assumed but not proven.

In figure 7 the tensile strength of the single setting series is given. There is a sharp decline in both the high and low series due to reprocessing. The difference between the first and last series is about 26 % while the scattering of the values is very low. Nevertheless, no difference in tensile strength can be detected between the low and high single setting series.

The specimens illustrated in figure 8 display a rise in elongation at break over the five processing cycles (12 %).

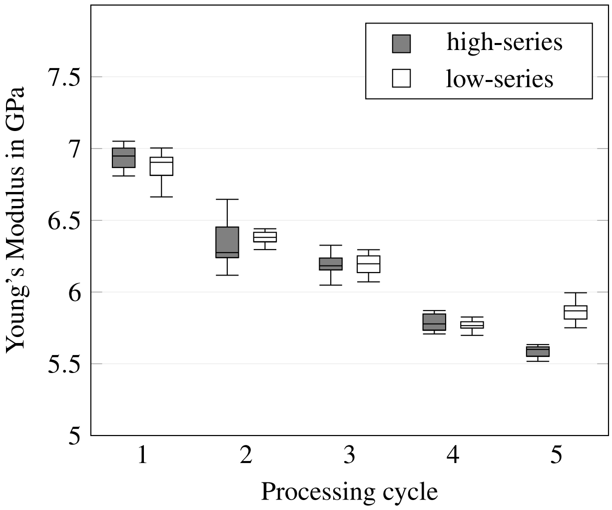

Fig. 6. Young's Modulus values for the five processing cycles for the single setting series (high, low)

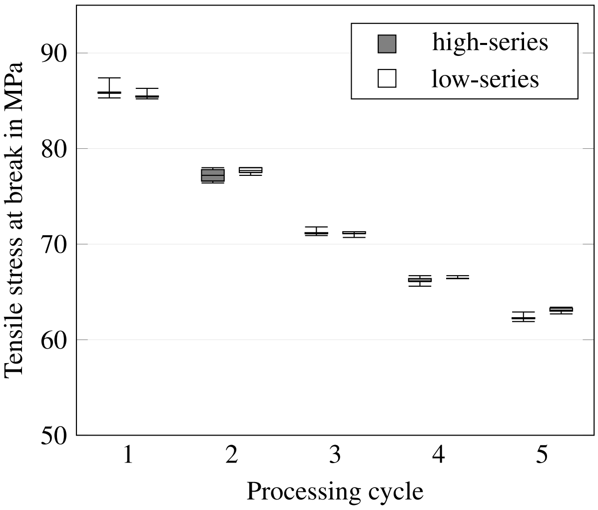

Fig. 7. Tensile stress at break for the five processing cycles for the single setting series (high, low)

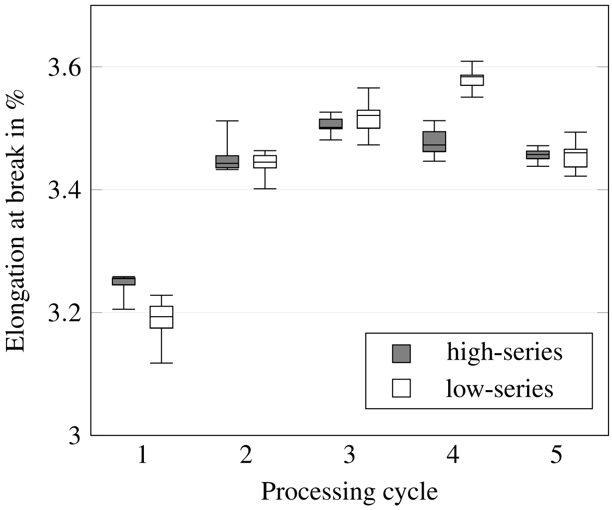

Fig. 8. Elongation at break values for the five processing cycles for the single setting series (high, low)

3.3.2. *Impact test*

Since the results of the Charpy impact tests were obtained from production reserve samples, the measurements allow only a first orientation. Figure 9 shows the impact strengths of the single setting series plotted against the processing cycles. Despite the rare data, it can be clearly seen that the impact strength is reduced as a result of repeated processing. This assessment is also reflected in the literature. [8]

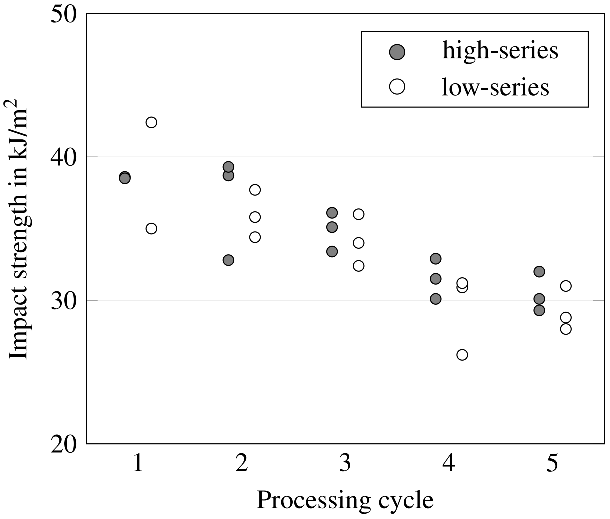

Fig. 9. Impact strength for the five processing cycles for the single setting series (high, low)

3.3.3. *Statistical evaluation of experiments*

For statistical evaluation of the single setting series, the processing cycle, the screw speeds, and the resulting material properties including their standard deviations were analysed via JMP. The results show, that for the single setting series the number of processing cycles has a significant correlation with the material properties. In contrast, screw speed shows no statistically significant effect on material properties in the single setting series. This means that the differences between the different screw speeds can be recognized in individual material properties, but they are not statistically reliable (cf. number-averaged fiber length in Fig. 4). In addition, the mixed setting series were statistically analysed. First, the number of high or low screw speed settings in the material history was investigated by introducing it as a separate variable factor. The fit least square model was then used to determine the P-Values. The results show, that no correlations are identified between the number of high or low screw speed setting and the material properties. Since there was no correlation between the number of settings in all five cycles, it is concluded that the order of the settings is negligible too.

4. Conclusion

The properties of short glass fiber reinforced thermoplastics are influenced by the injection molding process settings. This paper investigates if this influence can also be observed about multiple life cycles of the material. A glass fiber reinforced PP-GF30 was mechanically recycled up to five times (A.1). Two different screw speeds (high, low) were set for the injection molding process in each cycle. This allowed 47 different processing histories to be generated. The mechanical, rheological and morphological properties of the manufactured specimens were investigated. The studies indicate that material degradation is mainly caused by the number of reprocessing cycles. As a result of the reprocessing cycles, the fiber length distribution of the PP-GF30 shifted, leading to an increase of short fibers and a decrease of long fibers. After undergoing five processing cycles, the number-averaged fiber length has reduced by approximately 40 %. At the same time tensile strength (26 %) and Young's Modulus (16 %) decrease over the five processing cycles, while the elongation at break slightly increases (12 %). Along with fiber shortening, there is a sharp reduction in the MVR of the material (50 %). The resulting increase in flowability is also evident in the processing characteristics. The required injection pressures are significantly lower at higher reprocessing cycles. Moreover, the screw speed setting affects the injection pressure also in the single cycles. High screw speeds lead to lower injection pressures due to the shear thinning that occurs as a result of increased dissipation. The investigations also show that the influence of screw speed is not statistically significant. There was no correlation observed between the repetition or sequence of the high/low screw speed setting on the evaluated material properties. Summarizing it can be stated that the degradation behavior is highly influenced by the number of reprocessing cycles. The knowledge of the number of previous processing cycles is much more relevant to estimate the property degradation than the screw speed setting. The current investigation can help enhancing the understanding of cross life cycle relationships in injection molding materials. It offers a test methodology for future investigations on other influencing variables in the manufacturing, use, and end-of-life stage of a typical lightweight engineering material.

Acknowledgements

The project DiDe4Rec is funded by the German Federal Ministry of Economics and Climate Protection (BMWK) as part of the Technologietransfer-Programm Leichtbau (TTP-LB) initiative under the funding code 03LB3047.

Appendix A. Design of experiment

Table A.1. Excerpt from the used custom partial factorial design of experiment, ”−” = low screw speed, ”+” = high screw speed, * = single setting, others = mixed setting

Series	1. cycle	2. cycle	3. cycle	4. cycle	5. cycle
1	−	+	+	+	+
2	+	−	+	+	+
3*	−	−	−	−	−
4*	+	+	+	+	+

References

[1] R. Kupfer, L. Schilling, S. Spitzer, M. Zichner, M. Gude, Neutral lightweight engineering: a holistic approach towards sustainability driven engineering, Discover Sustainability 3 (1) (2022). `doi:10.1007/s43621-022-00084-9`.

[2] B. Klein, T. Gänsicke, Leichtbau-Konstruktion: Dimensionierung, Strukturen, Werkstoffe und Gestaltung, 11th Edition, Springer Fachmedien Wiesbaden, Wiesbaden, 2019. `doi:10.1007/978-3-658-26846-6`.

[3] F. Del Pero, M. Delogu, M. Pierini, The effect of lightweighting in automotive lca perspective: Estimation of mass-induced fuel consumption reduction for gasoline turbocharged vehicles, Journal of Cleaner Production 154 (2017) 566–577. `doi:10.1016/j.jclepro.2017.04.013`.
URL `https://www.sciencedirect.com/science/article/pii/S0959652617307096`

[4] Ali Hallal, Ahmed Elmarakbi, Ali Shaito, Hicham El-Hage, Advanced composite materials for automotive applications - 2013 - elmarakbi: Struktural integrity and crashworthiness.
URL `https://doi.org/10.1002/9781118535288.ch1`

[5] A. E. Schwarz, T. N. Ligthart, D. Godoi Bizarro, P. de Wild, B. Vreugdenhil, T. van Harmelen, Plastic recycling in a circular economy; determining environmental performance through an lca matrix model approach. `doi:10.1016/j.wasman.2020.12.020`.
URL `https://doi.org/10.1016/j.wasman.2020.12.020`

[6] Silvia Cecchel, Materials and technologies for lightweighting of structural parts for automotive applications.
URL `https://www.jstor.org/stable/10.2307/27033986`

[7] C. Hopmann, W. Michaeli, Einführung in die Kunststoffverarbeitung, 8th Edition, Carl Hanser Verlag, München, 2017.

[8] N. Rudolph, R. Kiesel, C. Aumnate, Einführung Kunststoffrecycling: Ökonomische, ökologische und technische Aspekte der Kunststoffabfallverwertung, Hanser eLibrary, Hanser, München, 2020. `doi:10.3139/9783446461284`.

[9] E. Baur, G. Harsch, M. Moneke, Werkstoffführer Kunststoffe: Eigenschaften - Prüfungen - Kennwerte, 11th Edition, Hanser, München, 2019.

[10] G. Oliveux, L. O. Dandy, G. A. Leeke, Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting properties. `doi:10.1016/j.pmatsci.2015.01.004`.

[11] L. Miller, K. Soulliere, S. Sawyer-Beaulieu, S. Tseng, E. Tam, Challenges and alternatives to plastics recycling in the automotive sector. `doi:10.3390/ma7085883`.
URL `doi:10.3390/ma7085883`

[12] V. A. González-González, G. Neira-Velázquez, J. L. Angulo-Sánchez, Polypropylene chain scissions and molecular weight changes in multiple extrusion, Vol. 60, pp. 33–42. `doi:10.1016/S0141-3910(96)00233-9`.

[13] G. Colucci, H. Simon, D. Roncato, B. Martorana, C. Badini, Effect of recycling on polypropylene composites reinforced with glass fibres, Vol. 30, pp. 707–723. `doi:10.1177/0892705715610407`.

[14] V. B. Gupta, Some studies on glass fiber–reinforced polypropylene. part i: Reduction in fiber length during processing.

[15] F. Gu, P. Hall, N. J. Miles, Q. Ding, T. Wu, Improvement of mechanical properties of recycled plastic blends via optimizing processing parameters using the taguchi method and principal component analysis, Vol. 62, pp. 189–198. `doi:10.1016/j.matdes.2014.05.013`.

[16] K. Brast, Processing of long-fibre reinforced thermoplastics using the direct strand-deposition process, Dissertation, Rheinisch-Westfälischen Technischen Hochschule Aachen, Aachen (2001). `doi:10.1016/S1464-391X(01)80187-2`.

[17] E. Kuram, Z. M. Sahin, B. Ozcelik, F. Yilmaz, M. M. Medar, Recyclability of polyethylene/polypropylene binary blends and enhancement of their mechanical properties by reinforcement with glass fiber, Vol. 53, pp. 1035–1046. `doi:10.1080/03602559.2014.886066`.

[18] Y. Guo, C. I. Chung, Dependence of melt temperature on screw speed and size in extrusion, in: Polymer Engineering & Science, Vol. Volume 29, pp. 415–419.
URL `https://doi.org/10.1002/pen.760290610`