*Technologies for Lightweight Structures* 5(1) (2021), pp. 61-68
Special issue: 5th International MERGE Technologies Conference (IMTC),
1st–2nd December 2021, Chemnitz

# Investigation of material degradation during multiple recycling loops of a glass fiber reinforced polypropylene compound to evaluate life cycle analysis based on mechanical properties

Ilka Pfisterer[1)], Roman Rinberg[2)], Lothar Kroll[2)]

[1)] Department of Research and Development for Interior Components, ilka.pfisterer@daimler.com, Mercedes-Benz AG, Bela-Barenyi-Straße 1, 71059 Sindelfingen, Germany

[2)] Department of Lightweight Structures and Polymer Technology, roman.rinberg@mb.tu-chemnitz.de, lothar.kroll@mb.tu-chemnitz.de, Chemnitz University of Technology, Reichenhainer Straße 31/33, 09126 Chemnitz, Germany

## Keywords

$CO_2$ Emissions, Life Cycle Analysis, Lightweight, Material Degradation, Polypropylene, Recycling

## Abstract

In order to reduce the global warming potential of their vehicles, automotive manufacturers are increasingly striving to use recyclates. However, recyclates often have weaker mechanical properties than comparable virgin polymers. Structurally, the weaker material properties can be compensated by an additional material effort. In semi-structural components of the vehicle interior, the bending stiffness is particularly important, which can be increased by a higher wall thickness to compensate for poorer mechanical properties, leading to higher component weights. The question is to what extent recyclates with poorer mechanical properties than virgin polymer result in $CO_2$-reductions in the overall life cycle.

In this work, long glass fiber reinforced polypropylene is recycled several times and the mechanical properties are determined. An LCA is carried out, based on bending stiffness as a functional unit to compare the advantages of recyclates with the disadvantages of higher component weights.

It turns out that in a vehicle with combustion engine only the first recycling loop results in a smaller GWP than the virgin polymer. For a vehicle with electric drive, this is the case for the second recycling loop.

## 1 Introduction and objective of the investigation

Due to the sustainability goals of the Paris Agreement and increasing social demands regarding waste prevention, the plastics industry is increasingly forced to rethink. In the automotive industry this is particularly true for the vehicle interior, as the majority of the components are manufactured from plastic. To achieve the imposed and self-imposed sustainability goals, OEMs develop different strategies. This includes the use of recycled materials and renewable raw materials, but also the reduction of material use by lightweight construction and load-optimized design.

Figure 1 shows the proportion of different types of plastic in the interior. The evaluation refers to components of cockpit, door trims and center console of premium vehicles. It can be seen that the plastic types PP, PC-ABS and PA with just over 90 % represent the largest proportion in the vehicle interior. The plastics are usually reinforced with glass or talc.

Digital Object Identifier: http://dx.doi.org/10.21935/tls.v5i1.160
www.lightweight-structures.de

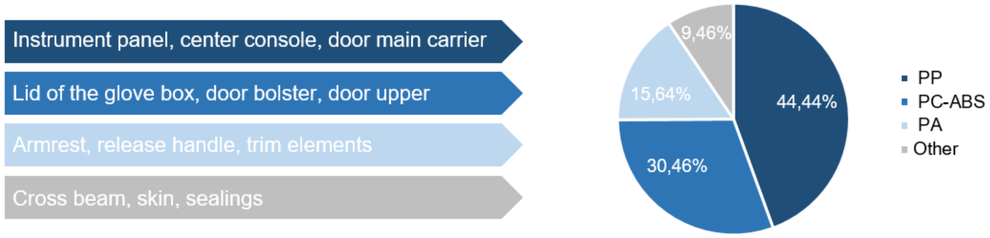


*Figure 1: Overview of the proportionate use of different types of plastic in automotive interiors determined from luxury class vehicles in the premium segment, taking into account the volume of production*

Components from long glass fiber reinforced PP (PP-LGF) are used in the vehicle interior in semi-structural components, such as the center console carrier. According to [1], PP-LGF is subject to a strong decrease in mechanical properties during the injection-moulding process as the fibers shorten under mechanical stress.

In terms of $CO_2$ reduction, the use of recyclate is especially worthwhile in material production, since the mass-related $CO_2$ emissions for recyclates are below those of virgin material. The obstacles for the use of the recyclate lay mainly in the large fluctuation range of material properties and lower mechanical properties. Lower mechanical properties can be compensated by higher dimensioning of the components. Since the use phase of the life cycle analysis (LCA) is weight-dependent, higher dimensioning results in higher $CO_2$ emissions. The question arises if the $CO_2$ emissions saved by using recyclates are overweighed by the necessary additional weight and therefore higher $CO_2$ emissions during the use phase.

In this work, a PP-LGF from series production is injection moulded and recycled several times. To compare the advantages of using recyclates and the disadvantages of higher dimensioning, bending stiffness is chosen as a functional unit (FU). As the bending stiffness is dependent on material properties and the component geometry, decreasing mechanical properties can be compensated by an increasing component thickness. An LCA is performed to ecologically evaluate the resulting $CO_2$ emissions. To compare different propulsion systems, the LCA is carried out with an internal combustion engine (ICE) and a battery electric drive (BEV). It should be noted that the internal combustion engine draws its energy entirely from fossil fuels, while the electric drive draws part of its energy from renewable sources due to the composition of the German electricity mix.

## 2 Materials and methods

For the test, a polypropylene reinforced with 18 % long glass fiber is used. The PP-LGF is made of the polypropylene PP 612MK10 EE from Sabic and the PP-Blend STAMAX 60YK270E with 60 % glass fiber content from Sabic. In order to investigate the degradation process over several recycling steps, the PP-LGF is injection molded ten times and recycled nine times, according to the test procedure in Figure 2. The recycling process includes one shredder and one compounding step. In the compounding step, no further fillers or additives are added and the material is granulated. The injection molding machine is of the type KraussMaffei KM-250-1400-C2, the compounding line is of the type Noris Plastic ZSC 25/40D. A shredder of the type Moreto Granulator GR3035 is used.

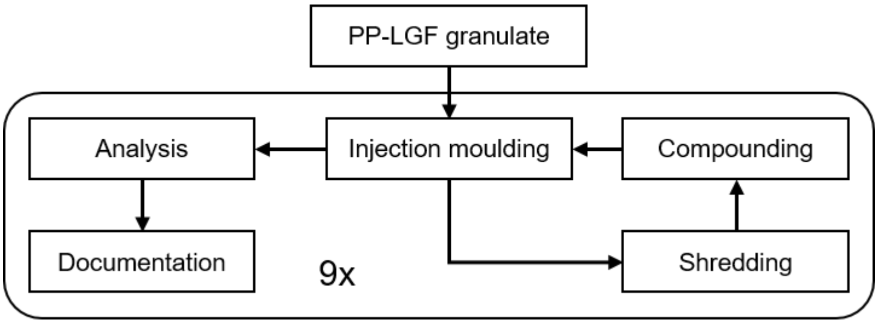


*Figure 2: Schematic experimental setup of recycling loop with PP-LGF granulate*

After injection moulding, some components are retained in each cycle for later analysis. Tensile tests and a fiber length analysis are carried out to investigate the degradation behavior and to determine the stability range. Bending tests are performed to carry out the LCA on the basis of bending stiffness.

## 2.1 Methods of testing for mechanical properties

Tensile test

The tensile test is carried out according to DIN EN ISO 527-2. The injection mould contains five tension specimen, which are used for testing and correspond to the specified dimensions according to DIN EN ISO 20753. The tensile test is carried out on a Zwick/Roell Z100, where five tensile specimen are tested per recycling loop. The tensile tests are carried out with a load cell of 10 kN and a test speed of 5 mm/min. During the tensile test, the applied force and the resulting sample length are continuously recorded. From the recorded values, stress-strain curves are determined and the modulus of elasticity is calculated.

Fiber length distribution

Since a shortening of the fibers can be assumed due to the mechanical stress during the injection molding and the compounding process, the average fiber length is measured. This results from the fiber length distribution, which is determined using the FASEP method of IDM Systems according to ISO/DIS 22314 on the basis of at least 1000 fibers. For the examination, three tests are carried out per test point.

Bending test

To analyze the bending stiffness of the material, three-point bending tests are carried out according to DIN EN ISO 178. The test is performed out on a Zwick/Roell 5.0 with a 1 kN load cell at a test speed of 2 mm/min. During the test, a stress-strain diagram is recorded and the bending modulus is calculated. For each recycling loop, five specimen are tested.

## 2.2 Method to carry out the Life Cycle Analysis

The LCA enables a systematic approach to calculate environmental impacts over the entire life cycle of a product. A distinction is made between different effect categories which, among other things, estimate the global warming potential (GWP), the acidification potential and toxicity potentials. The most common category of impact assessment is the GWP. To carry out the LCA, the GaBi software from Sphera Solutions, Inc. is used. The following steps are distinguished in the LCA calculation:

| | |
|---|---|
| Material manufacturing: | Production of the new material or recycling process in the case of recyclate use |
| Production process: | Production of components by injection moulding |
| Use phase: | Weight-related energy consumption with a mileage of 200,000 km with combustion or electric drive |
| End of life: | Incineration |

The impact assessment is carried out using the CML method according to DIN EN ISO 14040. The impact category is defined by the GWP over a period of 100 years (GWP 100). Standard values for polypropylene and glass fibers from the GaBi software database are used to balance the global warming potential of virgin material. The injection moulding process in the production step is calculated with an energy consumption of 1,2 kWh/kg, which is issued in the database of the GaBi software. The emissions of the use phase for an internal combustion engine $GWP_{ICE}$ are according to Eq. 1. The weight-related fuel reduction value $FRV$ of 0.15 l / (100kg · 100km) for compact cars according to the NEDC (New European Driving Cycle) [2] is assumed. The factor $m$ refers to the mass of the component, the mileage is indicated by $x$ and assumed to be 200,000 km. The emissions for the provision of the fuel $GWP_{well-tank}$ are assumed to be 0.46 kg $CO_2$-eq./l and the emissions of the fuel combustion $GWP_{tank-wheel}$ are assumed to be 2.33 kg $CO_2$-eq./l.

$$GWP_{ICE} = FRV_{(100kg,100km)} \cdot m \cdot x \cdot [GWP_{well-tank} + GWP_{tank-wheel}] \tag{1}$$

For the use phase of an electric vehicle $GWP_{BEV}$, an energy consumption $W_{(100kg,100km)}$ of 0.54 kWh/(100kg · 100km) is taken into account [3]. The mileage $x$ is also assumed to be 200,000 km. The factor $GWP_{electricity\ mix,D}$ represents the $CO_2$ emissions per unit of energy, which according to the German electricity mix amount to 0.547 kg $CO_2$-eq./kWh.

$$GWP_{BEV} = W_{(100kg,100km)} \cdot m \cdot x \cdot GWP_{electricity\ mix,D} \tag{2}$$

For incineration, the resulting emissions of the material are considered and a credit for the energy generated is awarded according to the German electricity mix. The data also comes from the GaBi software database.

## 3 Degradation of the PP-LGF over various recycling loops

The change in the mechanical properties of the PP-LGF due to the recycling loops is investigated on the basis of tensile tests. The tensile modulus allows an initial analysis of the mechanical properties and is a widely used characteristic value that describes the proportional relationship between stress and strain of the test specimen. In addition, elongation at break can be used to estimate the degradation progress of the polymer [4]. The tensile modulus determined over ten processing loops is shown in Figure 3, which indicates a sharp decrease within the first three loops. In loop 3–9, the tensile modulus decreases only slightly and stagnates at a level of 1500 MPa.

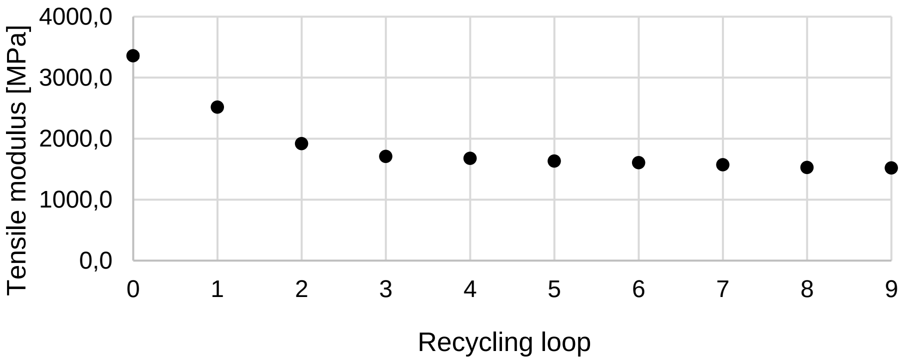


*Figure 3: Representation of the tensile modulus of the PP-LGF samples over ten processing loops (determined from tensile tests according to DIN EN ISO 527-2)*

The reason for the decrease of the tensile modulus is seen in the decrease of fiber length due to mechanical stress during compounding, granulating and injection moulding. Figure 4 shows a significant drop in fiber length within the first three recycling loops.

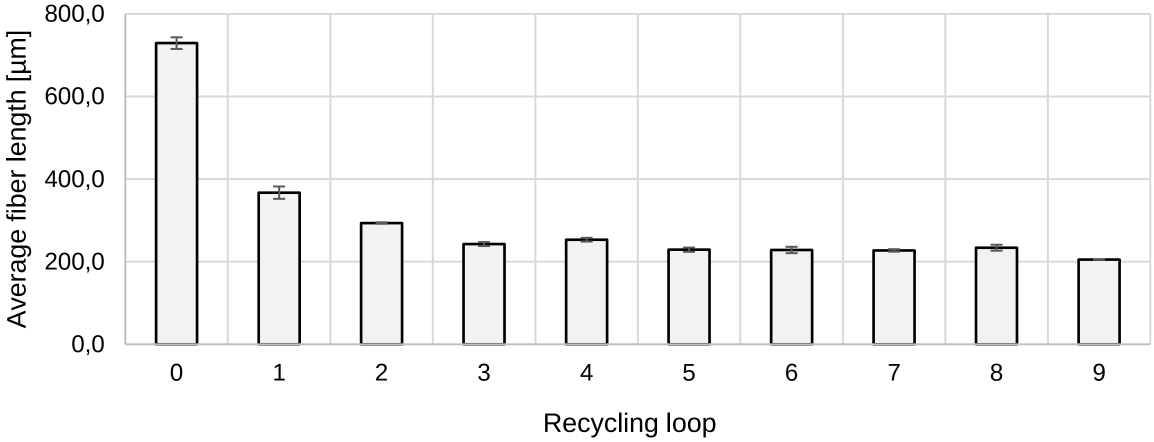


*Figure 4: Average fiber-length by number-average measured with the FASEP-method over ten processing loops*

The fiber length continues to decrease between cycle 3 and 9, but only to a small extent. The decreasing course of the fiber lengths over the recycling loops corresponds to the characteristics of the decreasing tensile modulus. With a length of approx. 200 µm, the fibers are still to be described as short glass fibers in recycling loop 9 [5]. Microscopy images of the glass fibers from recycling loops 0 and 3 are shown in Figure 5.

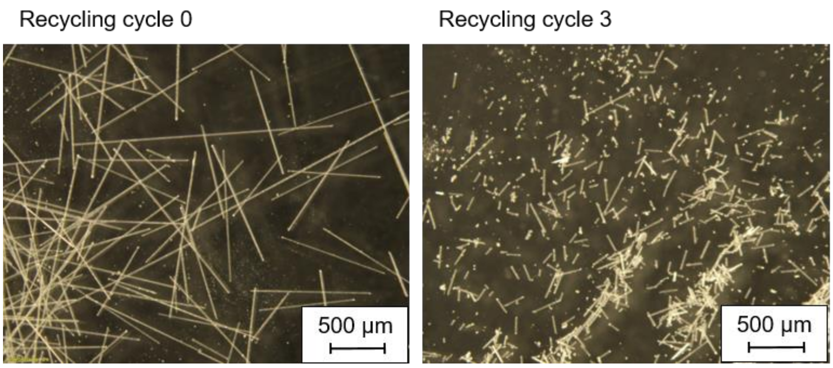


*Figure 5: Microscopy images of the glass fibers after ashing of the matrix to illustrate the fiber lengths*

The tensile modulus at loop 9 is comparatively high at 1500 MPa. According to the data sheet, the pure polypropylene has a tensile modulus of 1350 MPa. It can be assumed that the short glass fibers continue to have a reinforcing effect. A degradation of the polymer is not apparent from the measured values.

According to [4], the tensile modulus does not correlate with the degradation of the polymer. Instead, elongation at break is proposed as a characteristic indicator of age-related embrittlement, as it drops sharply as the plastic degradades progressively. Figure 7 shows the elongation at break over the recycling loops. The elongation at break increases within loop 1–3 and is accompanied by a large dispersion of measured values in recycling loop 5–9. The initial increase of elongation at break can be attributed to the shortening of the glass fibers and consequently the reduction of material stiffness. A decreasing elongation at break due to material embrittlement is not recognizable, even with constant fiber lengths between recycling loop 4 and 9. The large scattering band of the mechanical characteristics of the PP-LGF from cycle 5–9 cannot yet be explained with the analytical methods used. The use of PP-LGF from cycle 5–9 would result in great uncertainties and is not justifiable. Taking into account elongation at break as a characteristic indicator of material degradation, PP-LGF can be used up to recycling loop 4.

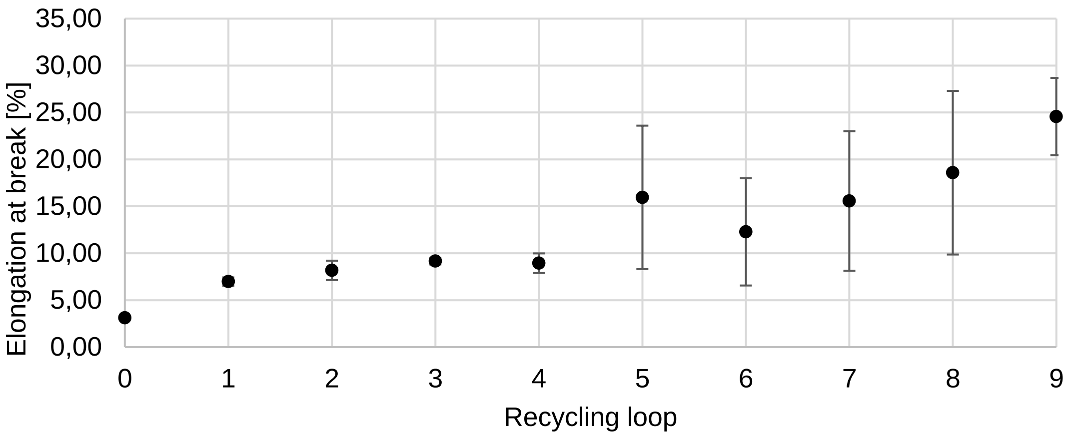


*Figure 6: Elongation at break of the PP-LGF samples over ten processing loops of the recycling simulation (determined from tensile tests according to DIN EN ISO 527-2)*

For semi-structural components in vehicle interiors that have to withstand high loads but are not crash-relevant, bending stiffness is a relevant parameter. The bending test determines the bending modulus shown in Figure 7.

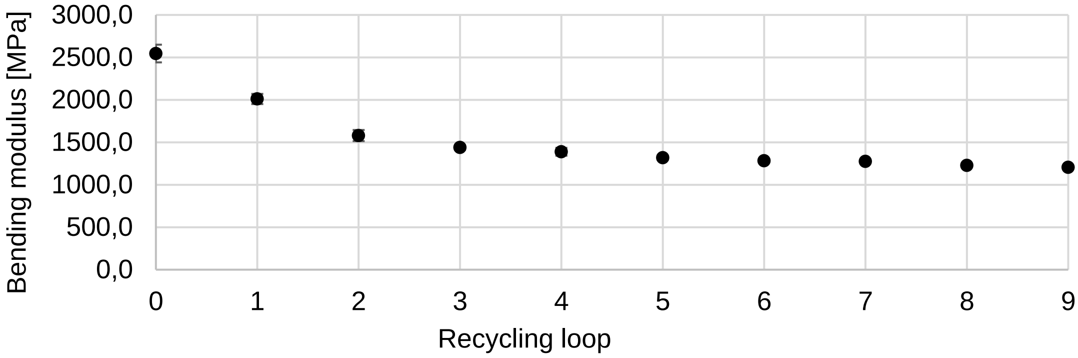


*Figure 7: Bending modulus of the PP-LGF samples over ten processing loops (determined from bending tests according to DIN EN ISO 178)*

Just as the tensile modulus, a sharp decrease is recognizable for the bending modulus up to recycling loop 3. From loop 4–9 only a small decrease is detected.

# 4 Creation of an LCA based on the bending modulus as functional unit

Bending stiffness is a significant parameter for interior components. Lower mechanical properties due to material degradation can be counteracted constructively by additional material.

The bending stiffness $D$ results from the bending modulus $E$ and the moment of inertia $I_y$ according to Eq. 3. For a plate geometry with a constant width $b$, the height $h$ can be adjusted to achieve a given bending stiffness. The bending stiffness of the specimen in recycling loop 0 is set as a reference value and serves as a functional unit for the LCA. To carry out the LCA, the necessary specimen height is analytically calculated, considering the bending modulus of chapter 3 for each recycling loop.

$$D = E \cdot I_y \quad mit \;\; I_y = \frac{b \cdot h^3}{12} \tag{3}$$

With the same sample width and length, the mass can be calculated according to Eq. 4.

$$m = \rho \cdot b \cdot h \cdot l \tag{4}$$

With further degradation of the PP-LGF and therefore poorer mechanical properties and a lower bending modulus, the necessary height to achieve a given bending stiffness increases, resulting in higher part weights.

Figure 8 shows the LCA with a combustion engine in the use phase. Cycle 0 serves as a reference using virgin material. The $CO_2$ values of polymer production are correspondingly high. According to the cut-off allocation, only the energy consumption of the recycling process is credited to the calculations of the recyclate for samples 1–3, which results in low $CO_2$ emissions for the material production phase. The use phase has the greatest impact on the LCA, which gives great importance to the component weight. Incineration at the end of life has only a small contribution to the overall $CO_2$ balance.

The recycling loops 2 and 3 have significantly higher $CO_2$ values in the use phase due to the additional material and thus the higher weight. For recycling loop 2 and 3, the lower $CO_2$ balance of material production is outweighed by the additional weight in the use phase and leads to higher $CO_2$ values in the overall balance. In total, only the sample from recycling loop 1 has lower $CO_2$ emissions than the sample containing virgin material.

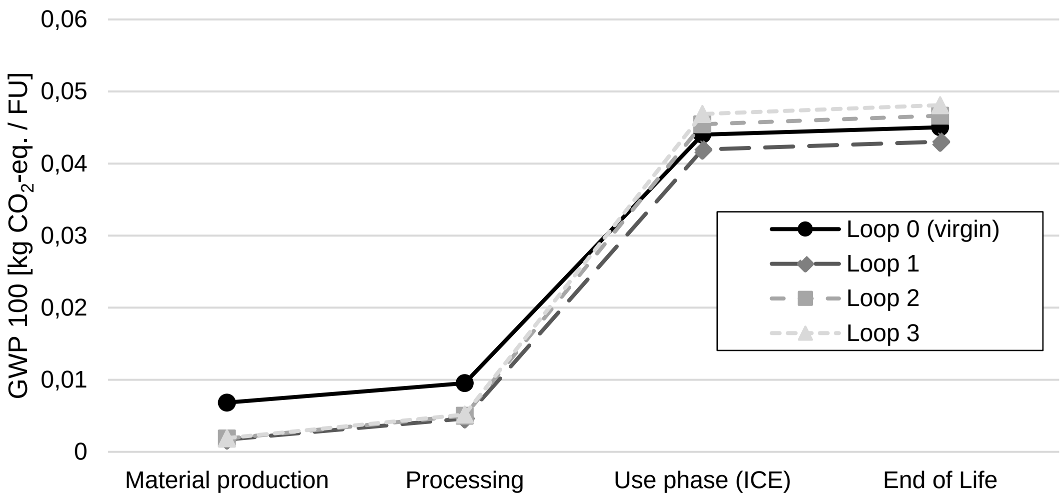


*Figure 8: Life cycle analysis based on bending stiffness as a functional unit over recycling loop 0–3 using a combustion engine during the use phase*

The LCA considering an electric drive in the use phase is shown in Figure 9. Compared to the results when using the combustion engine, a lower influence of the use phase and thus of the component weight on the LCA becomes clear. Only in recycling loop 3 do the increased $CO_2$ emissions due to the additional weight in the use phase exceed the $CO_2$ reduction due to the use of recyclates in material production leading to an overall increase in $CO_2$ emissions.

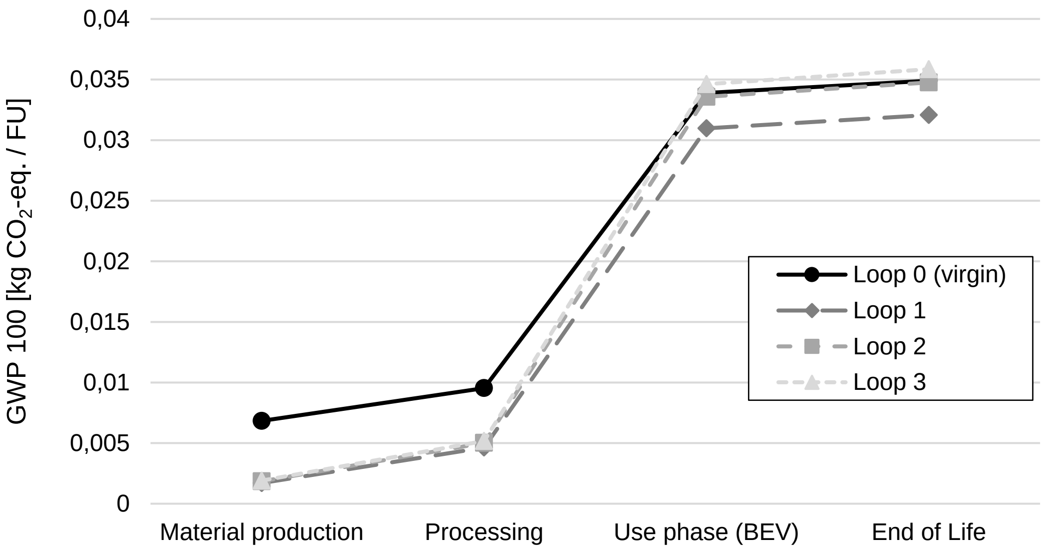


*Figure 9: Life cycle analysis based on bending stiffness as a functional unit over recycling loop 0–3 using an electric drive during the use phase*

## 5 Conclusion

In general, the decrease in mechanical properties of the examined PP-LGF is primarily due to the shortening of the glass fibers. Degradation of the polymer cannot be detected with the selected methods, but the material is subject to large fluctuations with regard to elongation at break from recycling loop 5 on. The use of mechanical characteristics as a functional unit in an LCA makes it possible to weigh up the use of recyclates, taking into account lightweight effects.

The LCA shows a strong dependence on the component weight, which is stronger pronounced in a vehicle with combustion engine than in a vehicle with electric drive. A slight weakening of the material can be compensated constructively by an additional material effort, while at the same time reducing the $CO_2$ balance. With a progressive weakening of the material, the additional material increasingly outweighs the advantages of the use of recyclates, which leads to a higher $CO_2$ balance. The statement is not applicable in general, as the LCA depends strongly on the type of plastic and the load case. In the present case of this work, the use of recyclate can be recommended from an ecological point of view.

The sudden wide dispersion of elongations at break from recycling loop 5 on cannot be fully explained. Possible reasons are a change in the crystal structure or changed shear stress ratios between fiber and matrix due to degraded avivage. In further studies, this phenomenon will be investigated in more detail.

## References

[1] Chen, T.; Mansfield, C; Ju, L.; Baird, D.: The influence of mechanical recycling on the properties of thermotropic liquid crystalline polymer and long glass fiber reinforced polypropylene. Composites Part B, 200 (2020), https://doi.org/10.1016/j.compositesb.2020.108316

[2] Koffler, C.; Rohde-Brandenburger, K.: On the calculation of fuel savings through lightweight design in automotive life cycle assessments. The International Journal of Life Cycle Assessment 15(1) (2009), pp. 128-135. http://dx.doi.org/10.1007/s11367-009-0127-z

[3] Rohde-Brandenburger, K.: Was bringen 100 kg Gewichtsreduzierung im Verbrauch? – eine physikalische Berechnung. In: Siebenpfeiffer, W. (Ed.), Leichtbau-Technologien im Automobilbau, ATZ/MTZ-Fachbuch. Springer Fachmedien Wiesbaden, 2014, pp. 179-189.

[4] Ehrenstein, G.; Pongratz, S.: Die Beständigkeit von Kunststoffen. Carl Hanser Verlag GmbH & Co. KG, 2007, p.151.