Direct Recycling of Cured Thermosets as Fillers in the Injection Molding Process

Sprue Recycling Conserves Resources

Thermoset molding compounds crosslink irreversibly during processing. Both the material from the sprue runner system, the process start-up and scrap components are typically sent for thermal recycling as waste. The granulation and direct recycling of this material in the injection molding process increases resource efficiency due to material recycling, but has an influence on the mechanical properties of the components. In the present study, the necessary parameters are investigated.

Recycling of sprue loss through shredding, blending with virgin material and direct recycling. © Fraunhofer ICT

Thermoset molding compounds have inherent disadvantages compared to thermoplastics, due to their irreversible chemical crosslinking during molding. Nevertheless, they have very good thermomechanical properties, excellent media resistance and high dimensional stability, which make them attractive for numerous applications. In addition, their price is extremely lucrative. In the "Lite$_2$Duro" research project, the Fraunhofer Institute for Chemical Technology (ICT), together with a consortium from industry and research, is investigating strategies to increase resource efficiency in the processing of thermoset molding compounds by directly recycling finely comminuted cured components.

The basic strategy of using cured molding compounds as fillers is well known from the literature. For example, the use of phenolic resin as a filler in polypropylene (PP) has been investigated [1, 2]. Other research groups have used cured bulk molding compound (BMC) as a filler in PP [3, 4]. Both studies refer to unreinforced PP as a reference, and consequently report significant improvements in the mechanical properties.

Where thermosets are used as matrix material, the granulated fillers are mixed with the virgin material in an upstream compounding process [5 to 8]. A common factor in all the studies is that only the basic mechanical properties of the recyclate-filled plastics are investigated. This current study also examines

- the processing properties in the injection molding process,
- the influence of the recyclate filler on the geometric properties of a precision component and
- the influence on the ecological footprint (LCA) of the component produced.

Two phenolic resin molding compounds from the manufacturer Vyncolit were used for the experimental tests:

Life cycle stages	Status quo (g CO_2-eq./adjustment ring)	New development (use of 8 wt.% of recycled material) (g CO_2-eq./adjustment ring)
Raw material acquisition	104	96
Production Baumgarten company	39	32
Distribution and use stage	1101	1101
End-of-life (without credits)	53	46
Credits	-29	-26
Total	1268	1258
Potential for reduction absolute (g CO_2-eq.)		**-11**
Potential for reduction relative (%)		**-0.8 %**

Table 1. Results of the LCA study according to the GWP 100 global warming potential.
Source: Fraunhofer ICT

- Vyncolit X7530, a PF-GF55, and
- Vyncolit X7320, a PF-(MD+GF)80.

The basic mechanical characterization was carried out on a pilot plant scale at Fraunhofer ICT using tensile test specimens. Subsequently, the dimensional stability and mechanical behavior under a realistic complex load were investigated for an oil pump adjustment ring (**Fig. 1**) under series production conditions, at Baumgarten automotive technics GmbH, Burbach, Germany.

Fig. 1. Oil pump adjustment ring made of Vyncolit X7320. © Fraunhofer ICT

Preparation and Injection Molding of the Recyclate

To obtain the recycled material, components and sprues from the two molding compounds were crushed using a single-blade crusher and a cutting mill. Two size classes of the recyclate were investigated:

- d < 250 µm and
- 250 µm < d < 1000 µm.

Immediately before injection molding, the recyclate was mixed in batches with the virgin phenolic resin in weight fractions between 4 and 20 %, homogenized by tumbling and fed into the material hopper of the injection molding machine.

The tensile test specimens were produced using a 6-cavity mold on a 30 mm screw diameter injection molding machine (type: Allrounder 320 C, manufacturer: Arburg). The oil pump adjustment ring was produced using KM 80/390 and KM 150/750 injection molding machines (manufacturer: Krauss-Maffei) in 1-cavity and 4-cavity molds (mold design and construction: Baumgarten automotive technics). In each case the transition to holding pressure was adjusted according to the injection path.

Impact Assessment Based on Global Warming Potential

The life cycle assessment (LCA) was carried out in accordance with ISO 14040 standards [9, 10] using the software GaBi [11] and the Gabi Professional database. The study determines the potential resource and greenhouse gas savings along the full life cycle of the oil pump adjustment ring, from manufacturing, to use, to disposal.

In order to demonstrate the effective contribution to recycling and circularity in terms of resource savings, a so-called "life cycle gap" analysis (LCGA) is also carried out in the evaluation phase [12]. The product system as well as the life cycle inventory of the LCA are available online [13]. The functional unit here is an adjustment ring for a controlled pendulum adjustment ring oil pump in a passenger car with an expected service life of ten years or 200,000 km. The impact assessment is based on the indicator of global warming potential (GWP) over a period of 100 years, measured in kilograms of carbon dioxide equivalents (kg CO_2-eq.), according to the "Environmental Footprint 2.0" method.

Mechanical Properties of the Component

The influence of the recyclate on the injection molding process is determined by investigating the maximum injection pressure as a function of the recyclate content, using the example of tensile test specimens produced from X7530 (**Fig. 2**). For both size classes, a clear increase in injection pressure was seen with increasing recyclate content. With the exception of the highest recyclate content of 20 wt.%, the recyclate particle size does not appear to have a significant impact on the injection pressure.

The tensile strength of the manufactured test specimens is determined according to DIN EN ISO 527–2 [14] (**Fig. 3**). A decrease in tensile strength of about 15 MPa is observed for recyclate contents between 4 and 12 wt.%. At higher contents of recyclate, the decrease in strength is more pronounced. Where the recyclate with a particle size of 250 µm < d < 1000 µm is »

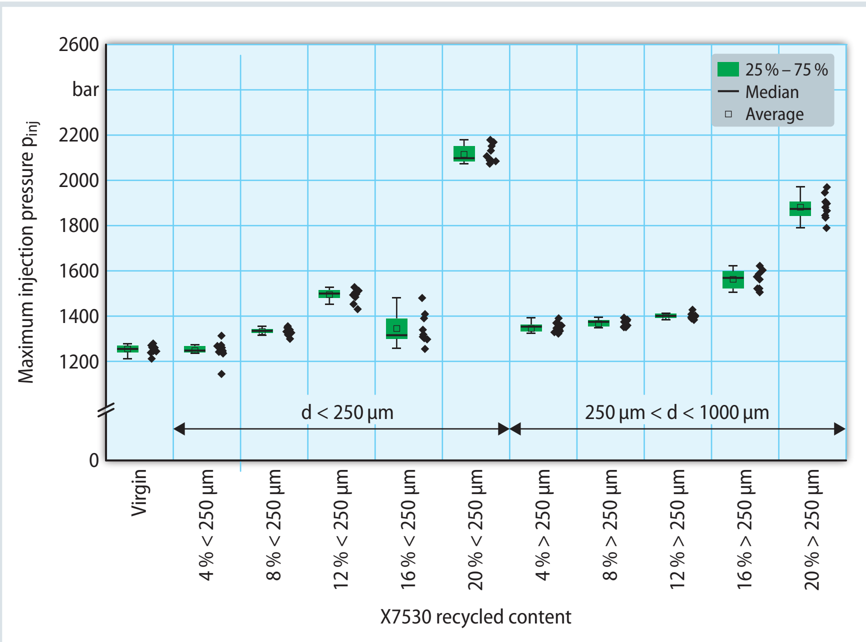

Fig. 2. Maximum injection pressure as a function of the recycled content for Vyncolit X7530.
Source: Fraunhofer ICT; graphic: © Hanser

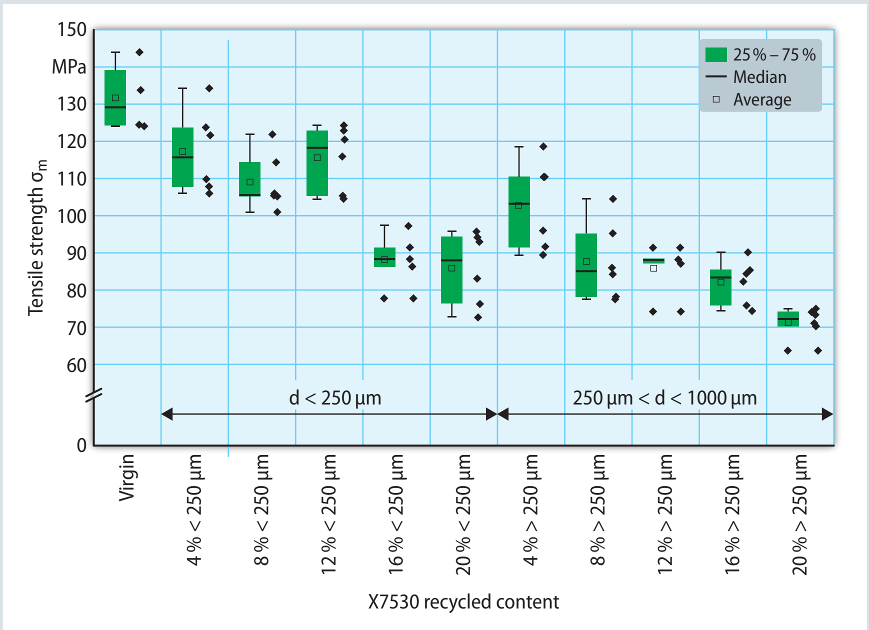

Fig. 3. Tensile strength as a function of recycled content for Vyncolit X7530. Source: Fraunhofer ICT; graphic: © Hanser

used, a sharp decrease in strength (25 MPa) occurs even at 4 wt.%. There is no discernible influence on the measurement scatter of strength for any size class or percentage of recyclate.

The fracture surfaces of X7530 virgin material and specimens with recyclate contents of 20 wt.% in both size classes (**Fig. 4**) were analyzed microscopically. Individual recyclate particles are hard to identify even at higher magnification. The skin and core layer-structure typical for phenolic resin molding compounds [15] is less pronounced at d > 250 µm, and the fracture surface is generally more jagged and irregular.

Comparable results were obtained for the molding compound X7320. Due to the significantly lower strength when using larger recycled particles (250 µm < d < 1000 µm), the injection molding tests for the oil pump adjustment ring were only carried out with the smaller size class (d < 250 µm). In injection molding, the required injection pressure also rose with increasing contents of recycled material. With unchanged injection molding parameters, mold filling with a recyclate content of 4 wt.% and above was only carried out during the injection phase of the process, and not during the holding pressure phase. This could be compensated by adjusting the holding pressure to the higher transition mass pressure.

Challenges in the Injection Molding Process

However, one factor emerged as problematic: the increasing tendency of the nozzle to clog due to material deposits building up on the inside. This phenomenon occurred at increasingly shorter intervals of 10 to 15 cycles for the high recyclate contents > 12 wt.% and could not be significantly mitigated despite numerous adjustments to the process parameters (including back pressure, screw speed, plasticizing temperature, mold temperature and injection profile).

The breaking load of the oil pump adjustment ring, determined in accordance with the requirements of series components, shows decreasing mechanical properties with increasing recyclate content, as was the case in the tensile strength tests. There is also a tendency towards higher measurement scattering. With an average breaking load of F = 1.6 kN, the material batch with a recycled content of 4 wt.% is still slightly above the breaking load of F = 1.5 kN, defined as the acceptable/ unacceptable limit.

Positive Contribution to Recycling

In contrast to the mechanical characterization, the size of the tolerance zone is not discernibly influenced by the recycled material content (**Fig. 5**). For all recyclate contents, a tolerance zone of s < 0.03 mm is achieved, which is well within the specification of s < 0.12 mm. There is a tendency for the diameter to decrease with increasing recyclate content. This is attributed to the higher cavity pressure that was required for complete mold filling in batches with a high recycled content [16].

The results of the LCA study according to the GWP 100 global warming potential show that the newly developed recycling process can effectively reduce greenhouse gas emissions by around 11 g CO_2-eq. per adjustment ring (**Table 1**), which corresponds to around 1 % of total emissions. From a circular economy perspective, the LCGA results (**Fig. 6**) show that the gap can be narrowed from the original 80 % to 77 %. The recycling of cured thermoset molding compounds from sprue distributors and from start-up and scrap components thus saves resources, and makes a positive contribution to closed-

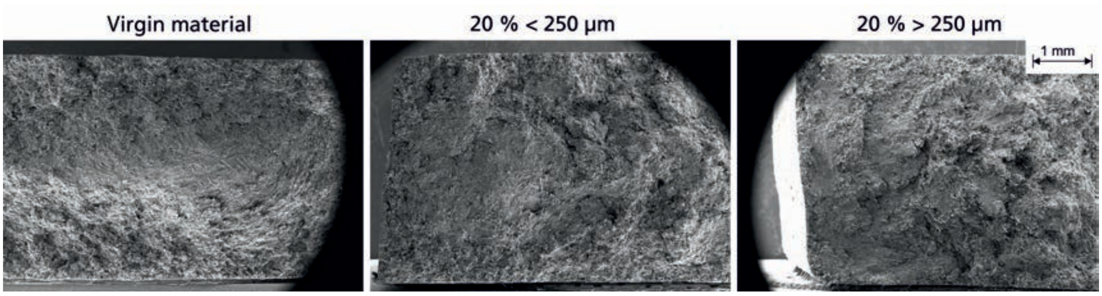

Fig. 4. Scanning electron micrographs of the fracture surfaces of test specimens made of Vyncolit X7530. © Fraunhofer ICT

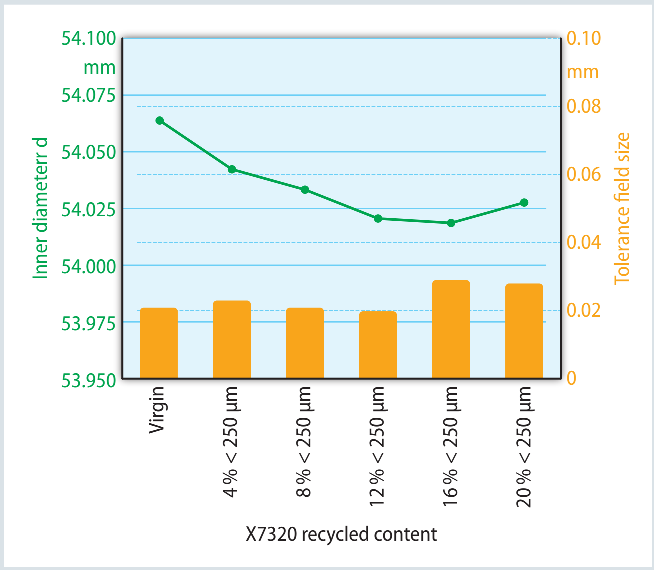

Fig. 5. Inner diameter and tolerance zone size of the oil pump adjustment ring as a function of the recycled content for Vyncolit X7320. Source: Fraunhofer ICT; graphic: © Hanser

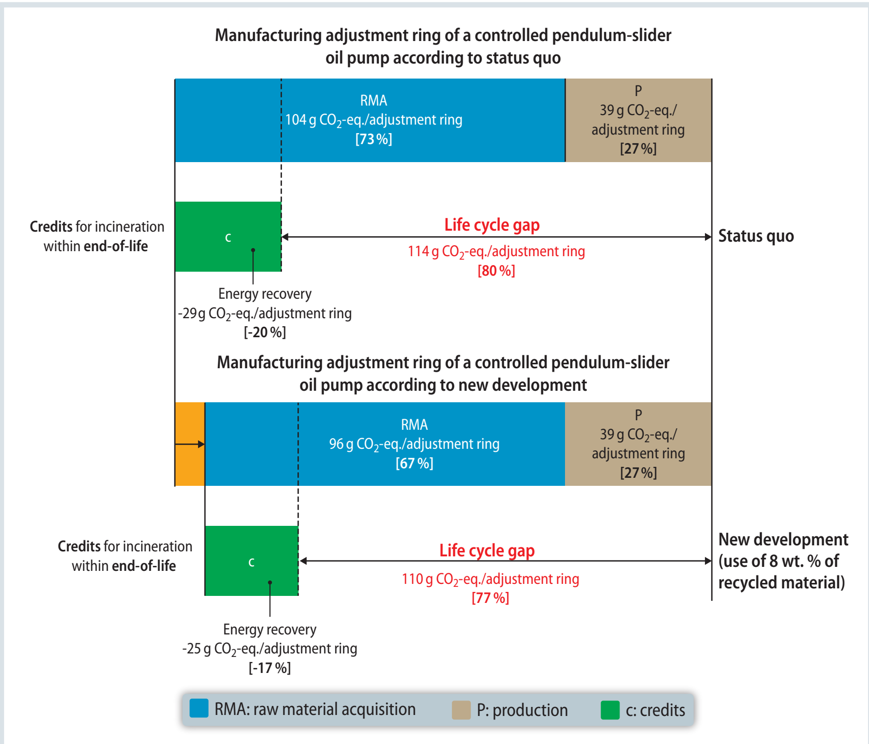

Fig. 6. Result of the "life cycle gap" analysis. The use of 8 wt.% recycled material reduces the gap between ideal recycling and real-life practice by 3 percentage points. Source: Fraunhofer ICT; graphic: © Hanser

loop recycling and circularity. At the same time, it is clear from the results that there is still significant potential for improvement.

Conclusion

The investigations show that it is possible to recycle cured thermoset molding compounds as finely ground fillers in the injection molding process. The feasible recycled content results from a trade-off between resource savings on the one hand and the loss of mechanical properties on the other. For the inorganically filled, glass fiber-reinforced phenolic resin molding compounds investigated in this study, the limit is a recycled content of approx. 8 wt.%. This proportion of recyclate results in a moderate loss of mechanical properties (-15 MPa tensile strength), an unchanged dimensional stability of the components and an increase in the maximum injection pressure of 100 bar. The GWP 100 global warming potential can be reduced by about 1 % and the gap in the cycle by about 3 %. In future studies, the aim will be to comminute the sprues directly at the injection molding machine, in order to achieve a closed cycle of the sprue material. ■

Info

Text

Robert Maertens, M.Sc., is a researcher in the Polymer Engineering Department at the Fraunhofer Institute for Chemical Technology (ICT), Pfinztal, Germany; robert.maertens@ict.fraunhofer.de
Michael Dieterle, M.Sc., is a researcher in the Environmental Engineering Department at Fraunhofer ICT.
Tayla Herrmann, B.Sc., is a research assistant in the same department.
Dipl.-Wirt.-Ing. Jan Hirz is Managing Director of Baumgarten automotive technics GmbH, Burbach, Germany; j.hirz@bat-duro.com
Dietmar Klaas is technical project manager at Baumgarten automotive technics.

Acknowledgments

The authors dedicate this publication to the memory of their colleague and former Institute Director of Fraunhofer ICT, **Prof. Dr. Peter Elsner,** and thank him for all his contributions and his friendly support. The work was carried out as part of the "Lite2Duro" project, which is funded by the German Federal Ministry of Economics and Climate Protection (BMWK) as part of the Lightweight Construction Technology Transfer Program based on a resolution of the German Bundestag (funding code 03LB3015C).

Service

More information about the project partners Fraunhofer ICT and Baumgarten:
www.ict.fraunhofer.de/en
bat-duro.com

References & Digital Version

You can find the list of references and a PDF file of the article at ***www.kunststoffe-international.com/archive***

German Version

Read the German version of the article in our magazine ***Kunststoffe*** or at ***www.kunststoffe.de***