Recyclates on the Test Bench

Incoming Goods Inspection of Recyclates Ensures Reject Minimization

Recyclates usually show altered properties compared to virgin plastics due to initial processing and influences during the product's life. This often leads to problems during processing. Especially in melt-expanding processes such as injection stretch blow molding, blow molding, film blow molding or thermoforming a fast incoming goods inspection method for recyclates is important. The Institut für Kunststofftechnik (IKT) at the University of Stuttgart, Germany, has developed a new test method for this purpose.

The responsible use of resources and raw materials is becoming increasingly important these days and poses challenges, for example in the disposal of waste and its return to the cycle. The Recycling Management Act (Kreislaufwirtschaftsgesetz) manages regulations on the disposal and recycling of waste since 1996 in Germany or at European level by EU Directive 2008/89/EC (Waste Framework Directive). The main purpose of those regulations are to conserve natural resources, to regulate the avoidance, recycling and disposal of waste, and to separate and sort recyclable materials. Plastics, especially thermoplastics, are particularly suitable for this purpose. [1]

In plastics technology, recyclates are therefore increasingly used for production of plastic products. In 2017, around 12.6 million t of plastic from primary raw materials and 1.76 million t from recyclates were processed in Germany. Plastic waste was almost completely recycled by the waste management industry. In the same year, 46 % of all plastic waste was materially recycled, 1 % of raw material recovery and 53 % thermally utilized [2]. Recycling rates are increasing every year and are expected to reach 63 % of materially recycled plastic in 2022, according to the German Federal Ministry for the Environment [3]. Plastics, including those from packaging, therefore not only have a "short life", but also several life cycles in a variety of applications. Therefore, it is essential to identify the mostly unknown material properties of recyclates before processing and/or post processing.

Influences of Recycling on the Material Properties

The processing of recyclates poses some challenges and difficulties. On the one hand, due to the shape of the recyclates, e.g. as a regrind, which can very often lead to a change in the processing process due to changes in bulk density depending on the regrind content and shape [4]. On the other hand, the material properties are often changed after previous processing and usage. This is manly caused by contaminations or mixing of the different types of plastics during the sorting process, but also by possible chemical, physical or mechanical aging and the associated degradation of the polymer chains during the life cycle [1, 5]. Reasons for contamination or degrada-

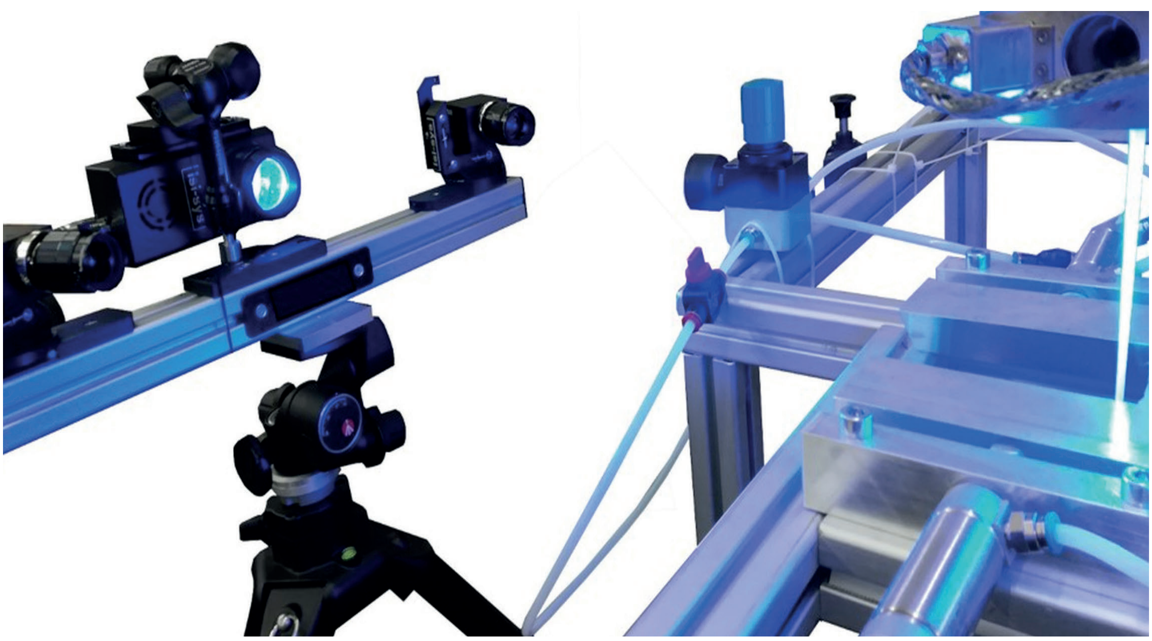

Fig. 1. The new and fast test method analyzes and identifies the melt extensibility of recyclates prior processing © IKT

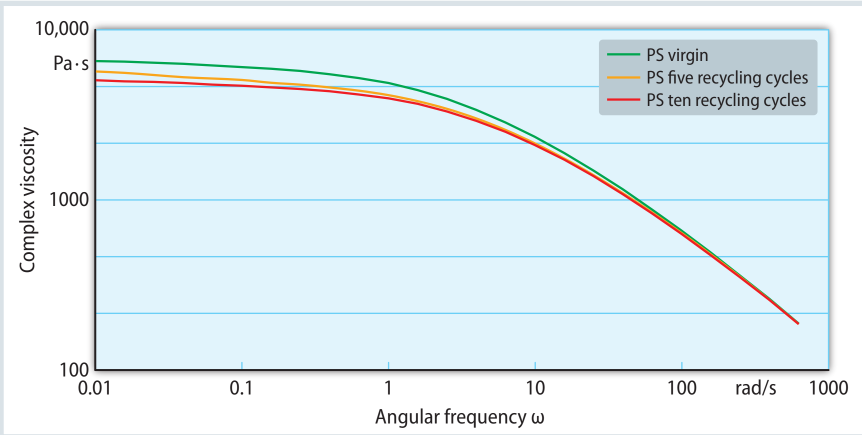

Fig. 2. Influence of recycling cycles on the shear viscosity of polystyrene at 220 °C Source: IKT, graphic: © Hanser

tion are the constantly growing amount of processed recyclates and the associated mixing during the recycling process, the different life cycles as well as applications (e.g. coloring, fillers, additives) and areas of usage (e.g. media influence) of the plastic products, which are ultimately returned to the processing process as mixed recyclates [5].

The effect of degradation can be illustrated by measuring the shear viscosity of polystyrene (type: PS 165N, Ineos Styrolution, Frankfurt am Main, Germany) and its recyclates at the IKT using a rotational rheometer (type: Discovery HR-2, TA Instruments, New Castle, DE/USA). The PS recyclate was produced at the IKT using a twin-screw extruder (type: ZSK 26, Coperion GmbH, Stuttgart, Germany). After the twin-screw extrusion process, the recyclates are then strand-granulated, dried and fed back into the twin-screw extruder. The changes in material properties of the recyclates can thus be analyzed as a function of the recycling cycles compared to virgin material. Looking at the curves, it is important to note that a double logarithmic plot is applied. Although, the zero shear viscosity (ω → 0 rad/s) seems similar, but they are quite different with 5082 Pa·s, 5719 Pa·s and 6612 Pa·s. Therefore, when the PS is recycled five and ten times, viscosity is found to be reduced by 13.5 % and 23.1 %, respectively compared to virgin PS (**Fig. 2**). The changes in viscosity not only have an influence on the pressure requirement of the process, but also on the mechanical properties of the produced component.

In order to minimize or even prevent rejects, it is therefore essential to analyze and identify the material properties of the recyclates before processing by means of a suitable incoming goods inspection. Established methods such as melt flow index measurement, tensile tests or infrared spectroscopy already exist for this purpose. However, these tests are not suitable to directly test the elongational properties of the melt of recycled plastics. Those properties are particularly important for processes such as injection stretch blow molding, film blow molding, blow molding or thermoforming.

Novel Test Method for Recyclates at the IKT

To determine the elongational properties of the recyclates without an additional production step of a semi-finished product or a test specimen, a novel test method was developed at the IKT and submitted for patenting to the Ger- »

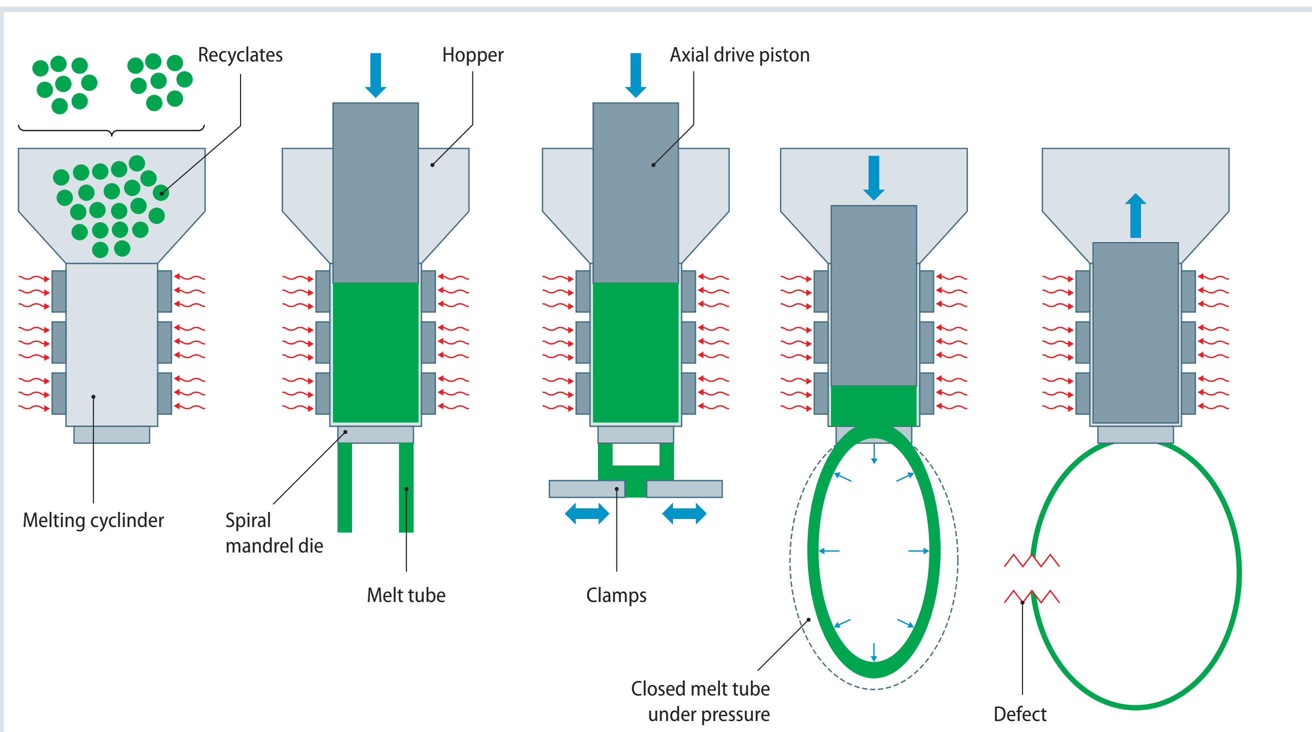

Fig. 3. Schematic sequence of the novel test method to analyze and identify the melt extensibility of recycled plastics Source: IKT, graphic: © Hanser

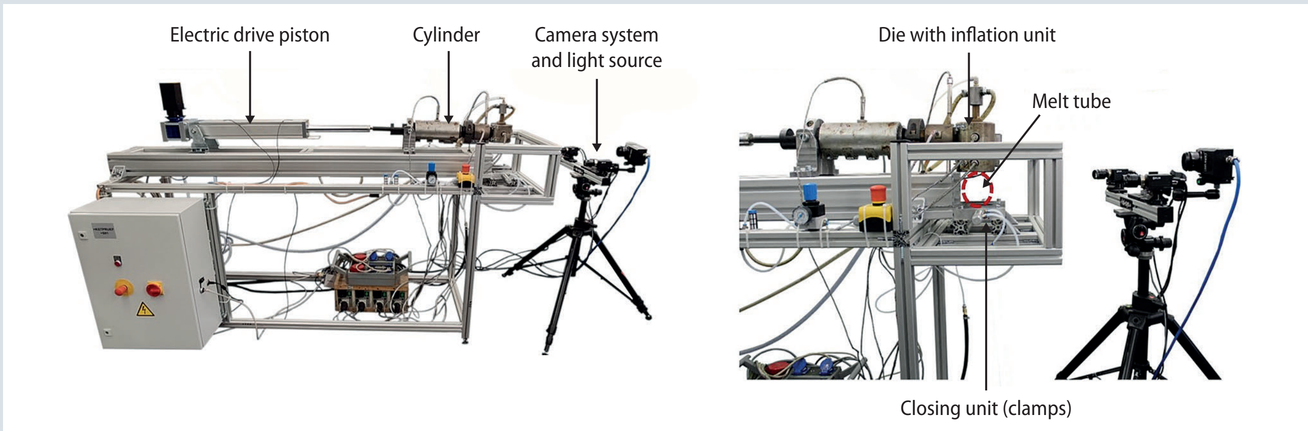

Fig. 4. The advantages of the new test method lie in the fast and simple handling of the test procedure Source: IKT, graphic: © Hanser

man Patent and Trademark Office, Munich, Germany (**Fig. 3**) [6]. The great advantages of the developed method are the simple handling of the test procedure, which runs as fast as a melt flow index measurement. Furthermore, real as well as close-to-process deformations (biaxial deformation) and velocities can be tested.

The test sequence involves the recyclates being fed to a melting cylinder by a hopper. The melt is then compressed by a piston and extruded through a spiral mandrel die to form a melt tube. The melt tube is closed after exiting the die. Subsequently, the sealed melt tube is inflated by means of compressed air until defect. During the inflation process of the melt tube, images are captured and recorded for monitoring and analysis of the inflation behavior. The evaluation of the images and image sequences over the inflation time is done by means of an image analysis software (Fiji, ImageJ). Thus, the biaxial deformation and the elongational properties of the recyclate melt can be analyzed as a function of temperature as well as of the applied air pressure and thus the deformation rate.

The test procedure is automated by the use of a microcontroller. An electrical driven piston with a maximum force of 15 kN is used to compress and convey the melt through the cylinder and the spiral mandrel die (**Fig. 4**). The pneumatic directional control valves are controlling the closing and inflation unit. Through a control signal from the microcontroller the system actuates the pneumatic pistons of the closing unit and starts the inflation process. The inflation conditions are adjustable by a pressure and flow controller before the start of the experiment. The five temperature zones along the cylinder and spiral mandrel die can be set and controlled separately. The resulting extrusion pressure is detected upstream of the spiral mandrel die. As a result, process control is significantly improved and irregularities are detected prior to measurement of the melt extensibility. Furthermore, the pressure measurement is directly coupled with the emergency shutdown of the piston, which ensures safe operations of the test procedure. The measurement sequence allows the extrusion speed, closure time, inflation time and camera recording time period of the test to be optimally adjusted to the recyclates tested. The geometry of the extruded tube can be modified by different wire guides and die inserts in the spiral mandrel die.

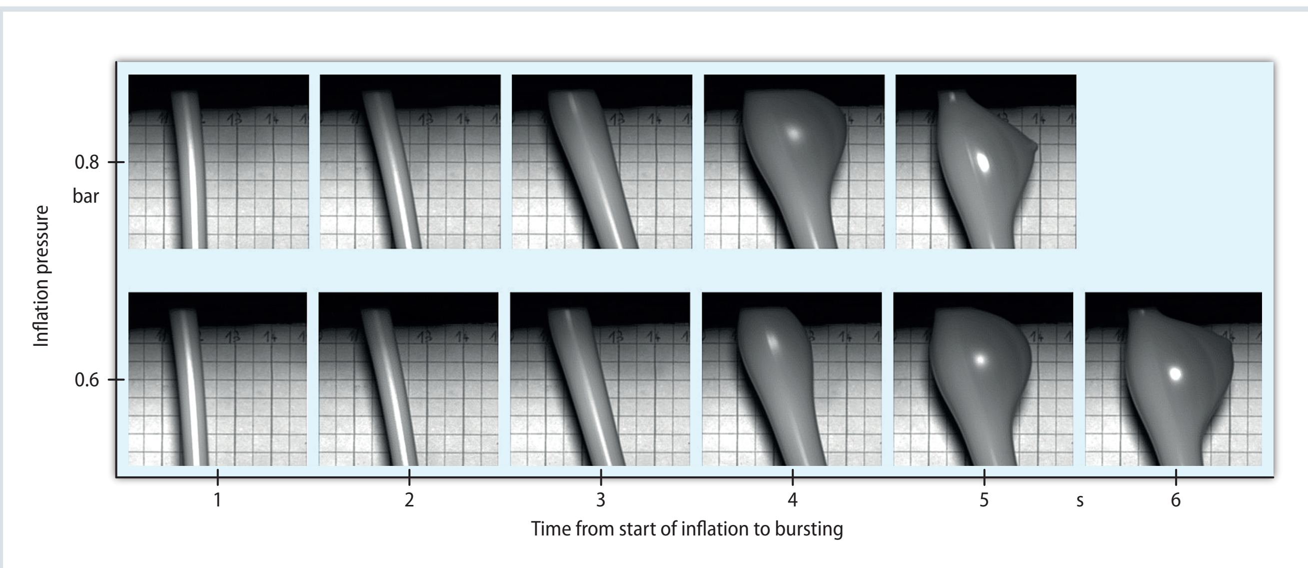

Fig. 5. Time sequence and development of the melt tube during inflation of PS virgin material at 240 °C Source: IKT, graphic: © Hanser

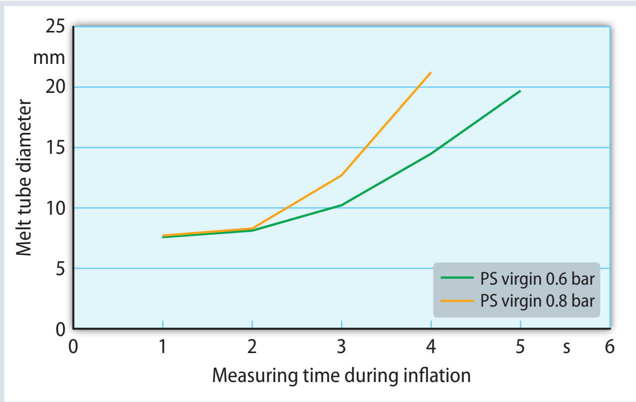

Fig. 6. Pressure dependency analysis of PS virgin material at 240 °C

Source: IKT, graphic: © Hanser

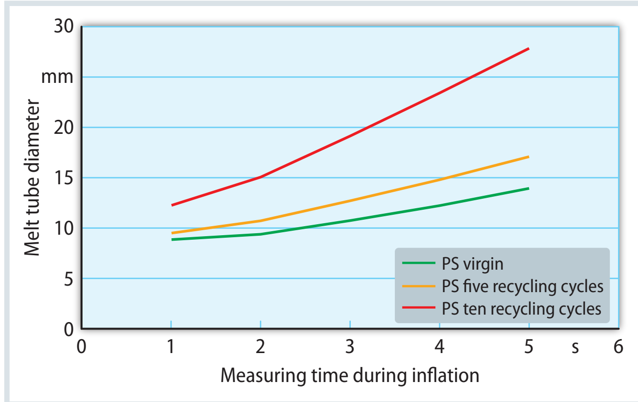

Fig. 7. Behavior of melt extensibility during inflation at 0.6 bar and 220 °C

Source: IKT, graphic: © Hanser

Measuring the Melt Extensibility of Recyclates

The melt extensibility measurements of the recyclates are carried out at an inner tube diameter of 4 mm and an outer diameter of 6 mm. The time sequence and development of the extruded melt tube during inflation up to defect or bursting was analyzed for PS virgin material at 240 °C and at inflation pressures of 0.6 bar and 0.8 bar.

Onwards after three seconds of testing, the pressure difference becomes directly visible within the images during inflation (**Fig. 5**). It can be seen that the measurement at 0.6 bar lasts longer until bursting or defect of the melt tube than at 0.8 bar. For quantification, the diameter of the melt tube is evaluated in each image sequence over the inflation process at the same distance from the outlet of the die. The point of evaluation can be chosen freely. However, all images should be analyzed at the same point from the outlet of the die, so that the different experiments are always evaluated at the same point and are thus comparable. The comparison of the inflation curves as a function of pressure is shown in a graphic (**Fig. 6**). Due to the higher inflation pressure, the melt tube is inflated faster and thus also fails earlier compared to the inflation pressure of 0.6 bar. The images at 5 and 6 s are not evaluated for 0.8 bar and 0.6 bar, respectively, because the melt tube had already burst open by then. Since the measurements are performed on virgin material and are corresponding to the expected behavior, it can be assumed that the measurement procedure is correct.

The experiments with recyclates are carried out at constant process parameters. The relationship between the reduction in viscosity (**see Fig. 2**) and the associated reduction in the internal cohesion of the polymer chains against an external force can be seen clearly for recyclates. This results in a stronger growth of the diameter of the recyclates melt tube compared to the measurements of virgin material or reference material. In order to classify measured recyclates and evaluate their influence on elongational properties, they always have to be compared with other recycled batches or virgin materials of the same grade. Recycled or virgin materials of a batch with a production of components without process or quality problems are suitable as references for analysis and comparison. Such a comparison was made for the virgin PS and its recyclates after being recycled five and ten times (**Fig. 7**). Similar to **Figure 2**, the influences of the recycling cycles can be seen. However, with the difference that the elongational behavior is analyzed.

If the measurement of the present recyclate or batch thus deviates from a reference curve, it can be assumed that process fluctuations may occur due to changed material properties.

Conclusion and Outlook

With the test method developed for analyzing the melt extensibility of recyclates, it is possible to quickly and easily test different recyclates in terms of incoming goods inspection for processes such as injection stretch blow molding, blow molding, film blow molding or thermoforming. By comparing to a "good" reference measurement, the suitability of the recyclate can be checked in a straightforward manner, as with an MFI measurement but for elongational properties.

In the next steps, the transferability of the results obtained so far with the novel test method will be further investigated and compared on the basis of blow molding and thermoforming experiments with various virgin materials and their recyclates. Furthermore, the use of a digital image correlation system is to be further considered for application. By that, strain deformations and strain rates during inflation can be examined over the whole melt tube. ■

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