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Editorial corner – a personal view

The growth and recyclability of thermoplastic polyurethanes

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Thermoplastic polyurethanes (TPUs) are one of the largest groups of thermoplastic elastomers. These TPUs are block copolymers whose molecular chains consist of hard and soft segments. Based on their properties, they form a bridge between thermoplastic polymers and chemically cross-linked elastomers because they can be processed and recycled in the molten state. At the same time, they are flexible, like rubbers (https://doi.org/10.1007/s12221-011-0857-y).
The first TPU was invented in the late 1950s and is considered the first thermoplastic elastomer in the modern sense. Since its introduction to the market in the 1960s, the popularity of this family of materials has remained unbroken. Nothing shows this better than that the TPUs market was estimated at USD 2.76 billion in 2022 and is projected to grow by nearly 7% by 2030.

TPUs are known for their flexibility, durability, and chemical resistance, making them suitable for use in industries as diverse as automotive, footwear, sports equipment, synthetic leather, electronics, and more. They are versatile materials with many applications, including films, sheets, fibers, tubes, and injection molded or 3D printed products. They replace conventional rubbers in many applications, such as tubes, shoe soles, or bicycle inner tubes (https://doi.org/10.3390/polym12091917).

As the amount of TPU used increases, so does the amount of waste that needs to be treated. The most common ways to treat TPU waste are landfilling, energy recovery, chemical or enzymatic degradation, and (thermo)mechanical recycling. In the spirit of the circular economy, it is essential to recycle these TPU-based products to reduce our environmental footprint.

Various recycled thermoplastic polyurethanes have recently become available from some raw material suppliers such as Covestro, Econa, or Gianeco. However, these recycled TPUs typically contain only post-industrial waste to ensure consistent quality. More and more manufacturers, including major brands (*e.g.*, Patagonia and Fairphone), are open to using this type of recycled material. This is partly due to legislation and partly due to social pressure.

Collecting large quantities of used TPU products from customers is still challenging. Separating TPU from post-consumer waste is complicated because, unlike polyethylene or polyethylene terephthalate, this family of materials does not have a specific resin identification code and is used in various products, often in combination with other materials. This process can change if manufacturers collect and recycle their TPU-based end-of-life products. Alternatively, used TPU can be collected through the distribution chain. A good example is the American startup HILOS, which offers its customers the opportunity to send shoes back at the end of life, enabling a circular future for footwear.

When recycling, it is essential to keep in mind that with each processing cycle, the polymers can undergo irreversible thermal, mechanical, and hydrolytic degradation, resulting in a reduction in molecular weight and a deterioration of properties. When the raw material is reprocessed and thus recycled, molecular weight decreases exponentially with the number of recycling cycles but is limited by an asymptote

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(https://doi.org/10.3390/polym12091917). Therefore, it is essential to know the number of reprocessing cycles the material can be exposed to (https://doi.org/10.1016/j.polymdegradstab.2022.109880). To increase their possible number of recycling cycles, it is always essential to choose the ideal, gentlest processing parameters (https://doi.org/10.1177/0095244314568691).

Nanni *et al*. studied the mechanical recycling of TPU parts of end-of-life ski boots to produce new boots fully circularly and investigated the effect of recycling cycles and the impact of the use of the boots on the recycled material. Several recycling iterations were performed, reducing tensile strength and elongation at break for TPU extracted from new boots after three recycling rounds. TPU from used boots showed comparable mechanical degradation after only two recycling steps (https://doi.org/10.1016/j.scp.2023.101059). This fact indicates that the materials degrade and deteriorate not only during recycling but also during use.

On the other hand, according to the literature, nanofillers, including layered silicates, can improve the mechanical and other properties (*e.g.*, gas barrier properties) of recycled materials. Various methods often modify the surface of fillers to create a better connection between the TPU and the filler; it can be plasma-treated or organically modified. Skrockiené *et al*. used oxygen, argon, and air plasma to modify bentonite clay to improve its affinity for recycled TPU. The unmodified bentonite shows poor reinforcing ability in the TPU matrix due to its low surface activity and difficulty in achieving complete platelet exfoliation. Modifying bentonite by plasma improves its dispersion in the matrix, thereby improving the mechanical properties of the blends (https://doi.org/10.1002/ppap.201500029).

As shown above, recycling thermoplastic polyurethanes is one path toward a circular economy. However, we still have much progress to make in the selective collection of this family of materials.