

Editorial corner – a personal view

Recyclable-by-design thermoset polymers and composites

A. Toldy*

Department of Polymer Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Műegyetem rkp. 3., H-1111 Budapest, Hungary

Thermosetting polymers form crosslinked structures with irreversible primary covalent bonds, so their production and processing, as well as their recycling, require different technologies than thermoplastic systems. Although thermosetting polymers are used in much smaller quantities (*e.g.* as engineering plastics) and have a much longer lifetime than thermoplastics, their recycling has become inevitable due to their increasing range and volume of use, higher price levels and the rapid growth in demand for carbon fibres. Despite the apparent difficulties, several recycling solutions exist, especially for fibre-reinforced composites. These methods can be divided into three groups: mechanical (where the composite waste is reduced to a smaller size and the material is recycled), thermal (where thermal energy is used to recover energy or material or both) and chemical (where the polymer matrix is decomposed by the addition of various compounds and both the matrix and the fibre reinforcement are recycled). From these, only mechanical recycling and pyrolysis (high-temperature anaerobic thermal decomposition) are currently available on an industrial scale, but some promising innovative concepts are aiming at recyclable-by-design thermoset polymers and composites. In 2011, Leibler and his colleagues introduced a new family of thermosetting materials, named vitrimers, which behave like crosslinked systems below the glass transition temperature but can be reshaped or even recycled similarly to thermoplastics above their topology freezing transition temperature (<https://doi.org/10.1126/science.1212648>). This thermally triggered reversible crosslinking is achieved via the dynamic rearrangement of covalent bonds in an associative manner

(<https://doi.org/10.1039/C5SC02223A>). Vitrimers containing dynamic covalent diketoenamine bonds could be depolymerised at low temperatures and short reaction times, enabling the chemical separation of the monomer and the additives as well (<https://doi.org/10.1038/s41557-019-0249-2>). In 2019, polyimine-based vitrimers were patented ([US20200247937](https://patents.google.com/patent/US20200247937)), which could be the first commercial vitrimers to replace epoxy resins in structural composites thanks to their high glass transition temperature. Another patented approach is applying amine-type curing agents specially designed for recycling that enable the hydrolysis of the crosslinked epoxy network under mildly acidic conditions. After the hydrolysis fibre reinforcement is recoverable in its original state, the dissolved matrix can be precipitated by alkaline neutralisation in the form of a thermoplastic epoxy ([WO2012071896](https://patents.google.com/patent/WO2012071896)). Alternatives to these innovative recyclable crosslinked systems are the already commercially available methacrylate-based thermoplastic systems in-situ radically polymerised with peroxides, which can be processed using thermosetting polymer technologies, provide similar mechanical properties and can be recycled by both chemical (depolymerisation) and mechanical methods ([FR2981652A1](https://patents.google.com/patent/FR2981652A1)).



Dr. Andrea Toldy
Member of Executive Editorial Board

*Corresponding author, e-mail: atoldy@mail.bme.hu
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