Combining the Best of Two Polymer Worlds

Thermoformable Acrylic Resin Systems for Large-Scale Production

Thermosets offer high mechanical performance and, thanks to their low initial viscosity, permit the efficient series manufacture of composite parts by resin transfer molding. The main advantage of thermoplastics is that they can be thermoformed, welded and recycled. These advantages are now combined in a newly developed thermoplastic acrylic resin system, which offers new possibilities for the production of fiber-reinforced composites with short cycle times.

Fig. 1. Advantages of the thermoplastic acrylic resin: The composites can be thermoformed and welded (© Arkema)

The large-scale production of fiber-reinforced polymer composites (FRPC), e.g. in automotive industry, is dominated by thermosets. Although thermoplastics offer various advantages such as weldability and thermoformability (**Fig. 1**), they are often clearly at a disadvantage regarding mechanical properties. They are even more disadvantageous concerning their processing behavior: The extremely high viscosity of the molten mass allows only very short flow paths in reinforcing fiber structures and therefore requires multiple semi-finished products to reach a final FRPC-component. On the other hand, thermosets can be injected in reinforcing fiber structures in a state of unlinked monomers having a near water-like viscosity allowing the resin to penetrate, or "wet-out" very long flow paths in a relatively short time. Consequently, nowadays there are very efficient, highly automated processes such as the resin transfer molding (RTM) that can be applied. In these processes the dry fiber structure is placed in a closed tool and impregnated with a thermoset resin system by pressurized injection.

Therefore, the target of current research on matrix polymers is to overcome the classical barrier between thermosets and thermoplastics in order to combine the advantages of both worlds [1]. In this context, the Institute for Composite Materials (IVW), Kaiserslautern, Germany, and the French chemical company Arkema work on a new generation of acrylic resin systems.

Thermoplastic Resin System for Composites

In the year 2014 the French chemical company brought liquid thermoplastic resins to the market under the trade name »

	Epoxy resin	Elium resin
Degree of crystallinity	crystalline	amorphous
Density [kg/dm^3]	> 1.16	1.18
Tensile modulus [MPa]	2800–3400	3100–3300
Tensile strength [MPa]	45–85	55–76
Flexural modulus [MPa]	2600–3600	3250
Flexural strength [MPa]	100–130	130
Elongation at break [%]	1.3–5.0	4–6

Table 1. Comparison of the mechanical properties of Epoxy and Elium resin (source: [4])

Fig. 2. Painted bus front end: The glass fiber composite was manufactured in cooperation with MVC Plasticos (© Arkema)

Elium. They provide mechanical properties comparable to epoxy resins (**Table 1**).

Since the resin system, unlike other thermoplastic resin systems, is liquid at room temperature (100–500 mPas), it can be processed with the same processes as epoxy resins, e.g. vacuum infusion, resin transfer molding (RTM), and wet compression molding.

The acrylic resin systems react to create a fully thermoplastic polymer via in-situ radical polymerization Thereby giving a classic resin-hardener combination which allows easy conversion to usage of this acrylic resin system. Due to the radical components a very short cycle time is achievable. Besides the already mentioned advantages, Elium, unlike sheet molding compound, does not contain any styrene, which clearly improves safety in the workplace. The thermoplastic composite-material, which was awarded with the 2013 JEC Innovation Award in the category "Thermoplastics", can be used to manufacture carbon-, glass- or natural fiber-reinforced composites (**Fig. 2**).

Concepts for Large-Scale Manufacturing

To exploit their full potential, the new matrix polymers have to be integrated in a holistic process concept as suggested in **Figure 3**. The first step is the cutting and stacking of the textile lay-up. Afterwards, the preforms can be brought into near-net-shape e.g. by binder-preforming. To investigate any potential obstacles when the new resin system is used in liquid composite molding, a joint project was conducted by IVW and Arkema, in which RTM plates were manufactured based on preforms with different binders. Thereby, it was proven that the acrylic resin is, from a process point of view, compatible with thermoplastic binder web as well as with epoxy-based thermoset powder binder. Also, for preforming the established standard processes can be used.

In a third step the impregnation takes place. For this the processes from RTM-family for thermoset resin systems in small and intermediate batch sizes are expected to provide the best performance. The target of the research project was also to manufacture thermoplastic FRPC parts in such a process. Thus, in a further study ex-

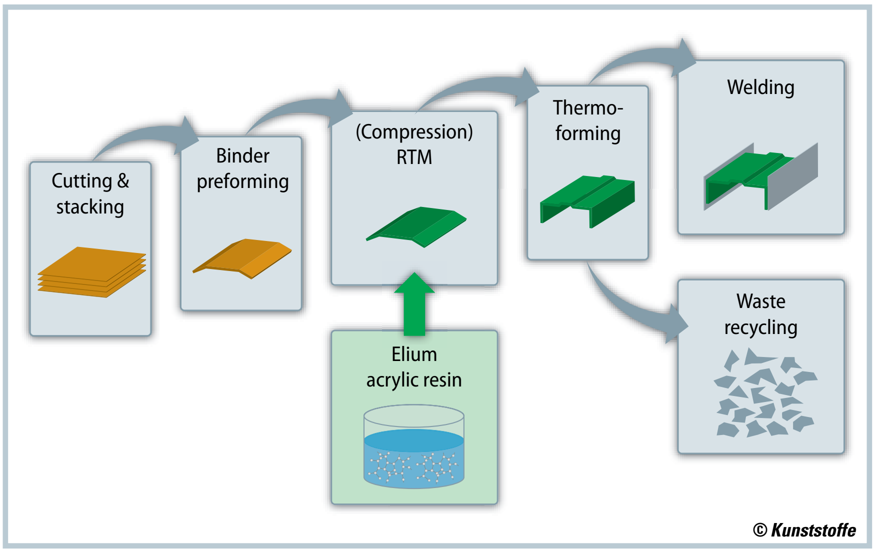

Fig. 3. Large-scale manufacturing: Process steps for thermoplastic resins. Also the recycling of waste as a press mass is possible (source: IVW)

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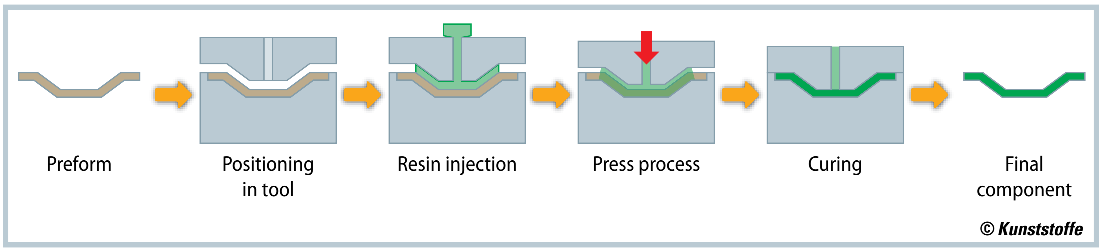

Fig. 4. Compression RTM: The process chain of the gap impregnation process (source: IVW)

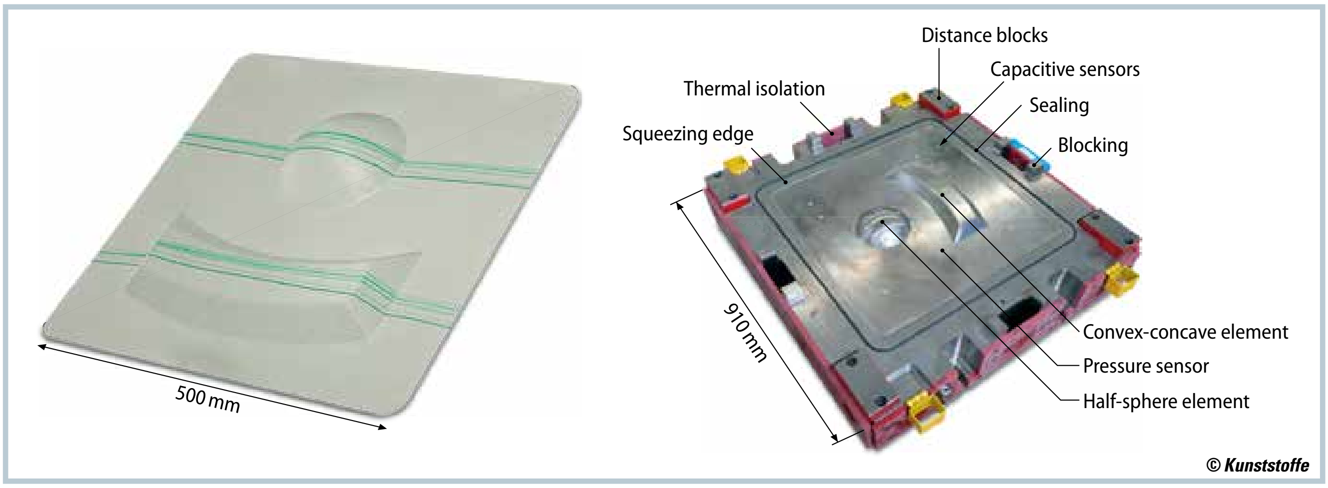

Fig. 5. 3D demonstrator part manufactured via compression RTM using the Elium resin (left) as well as the technology carrier tool and the tool carrier used for the manufacturing (source: IVW/Arkema)

emplary 3D-parts were manufactured within a compression RTM process (**Fig. 4**) in which the tool is not closed to the intended final thickness before injection, but the cavity height is initially slightly higher during injection. Therefore, the fiber volume content is strongly reduced, which allows higher flow velocities and a more efficient impregnation of the composite part. The cavity height during injection is raised so that there is a gap between upper tool and the preform. This allows the resin to flow over the preform. When sufficient filling is reached, the cavity is then closed to the final intended thickness, leading to full impregnation by pressing. Due to its high efficiency, especially for rather flat parts, the compression RTM is highly suitable for serial production.

Results

Figure 5 exemplarily shows a 3D demonstrator part which was manufactured by using the novel acrylic resin system, including the technology carrier tool and the tool carrier of IVW used within the study. Due to the low viscosity at room temperature (100 mPas), a full impregnation could be reached in short time. Depending on part complexity and process performance the cycle time can be optimally adjusted between a few minutes to several hours.

The time at which the tool is closed to the final intended fiber volume content is crucial for the compression RTM. In this context capacitive sensors, which were developed by IVW and further developed and sold by the Präzisionsmaschinenbau Bobertag GmbH, were used. They are implemented in the technology carrier tool and allow the fast and reliable detection of the flow front arrival at the sensors therefore allowing a "look" inside the closed steel tool [2]. This way, the tool can be automatically or manually closed as soon as the flow front arrives at certain points within the tool, in order to have maximum process reliability.

Besides the high efficiency of the compression RTM and similar processes, such as the wet compression molding, the process has a severe disadvantage: The restrictions concerning the geometry, which result from the closing process. The press process limits the complexity of the parts more or less to shell-like forms. Thus, despite their efficiency, these processes cannot always be applied. In this context, the acrylic resin systems offer unique advantages.

Thermoforming, Recycling, and Welding

Chemically the fully polymerized resin system is a thermoplastic and offers many of the advantages of this polymer group. Thus, a thermoforming step could follow the liquid composite molding (**Fig. 3**). The compression RTM could be used to manufacture a shell component (thermoform temperature is about 180 °C to 200 °C whereby the complex net-shape results from a subsequent thermoforming step. Also an additional welding process would be possible.

With regard to the growing importance of multi-material-design ("the right material at the right place") the need for corresponding joining technologies increases. The new acrylic resin systems allow welding and can therefore be integrated in new multi-material car body and chassis concepts. Considering the high material prices and the growing requirements for resource savings, the question of recyclability is also highly relevant. Thermoplastics offer advantages in this context, therefore Elium resin does as well. The waste occurring during trimming of the parts can be shredded and used again (e. g. as additional material for rib structures). ■