Cost efficient carbon fibre reinforced thermoplastics with in-situ polymerization of polyamide

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Abstract: Lightweight design has gained more and more relevance over the last decades. Especially in automotive industry it is of paramount importance to reduce weight and save fuel. At the same time the demand for safety and performance increases the components’ weight. To reach a trade-off between driving comfort and efficiency new lightweight materials have to be developed. One possible solution is the usage of carbon fibre reinforced thermoplastics (CFRTP) as a lightweight substitute material. In contrast to conventional carbon fibre reinforced plastics (CFRP), CFRTPs are cheaper and have a higher impact resistance. Furthermore they are characterized by hot forming ability, weldability and recyclability. However, the impregnation of the textile requires high pressure, because of the melted polymer’s high viscosity. A new innovative approach for CFRTP is the usage of in-situ polymerization with ε-caprolactam as matrix, which has a much lower viscosity and thus requires much lower pressure for impregnation and consolidation.

1. Introduction and Motivation
The need for lightweight structural materials has become increasingly important in the past years. Especially in automotive industry it is paramount to reduce weight. Due to enforcement of new emission regulations, rising fuel costs and the emerging electromobility manufacturers are forced to redesign their materials. At the same time higher safety standards, increasing demands in comforts and driving performance as well as space and quality arose. Thus, almost every new car generation shows a higher weight. This is depicted in the principle of the “weight spiral” (cf. Figure 1).

The demand for a cost-efficient high-performance lightweight material is given. In aerospace applications carbon fibre reinforced plastics are used to reduce the weight and therefore enable to build more efficient vehicles. CFRPs consist of two main components. These are the textile of reinforcement fibres and the surrounding matrix system. Nowadays the most common matrix systems are thermoset-based and could not enter the automotive market. This is mainly because of their long cycle time during production, moderate recyclability and reparability as well as high production costs.

Using thermoplastic materials like polyamide (PA) as matrix material is a promising new approach to overcome these drawbacks. In contrast to conventional thermost-based composites CFRTPs are recyclable, weldable and can be warm reshaped, thus increasing the profitability. CFRTPs are manufactured in the film stacking process. As the thermoplastic matrix material has a high viscosity, high pressures as well as high temperatures are necessary to completely impregnate the
textile. One option of reducing the temperature and pressure requirements is the usage of in-situ polymerization of the thermoplastic matrix.

![Figure 1. Reversing of „Weight Spiral“ [1,2]](image)

2. Advantages of carbon fibre reinforced thermoplastics

In contrast to conventional carbon fibre reinforced plastics, CFRTPs offer manifold advantages. One major advantage is the fast manufacturability as thermoplastics do not need long curing times for the chemical crosslinking reactions. Furthermore reasons to use thermoplastic resin systems are:

- Higher toughness and impact strength as compared to thermoset-based composites
- Hot forming ability and weldability ➤ joining of different components
- Good storage stability ➤ shelf life of intermediate materials is unlimited
- Recyclability
- Possibility of back injection

The future of global CFRTP market looks promising with opportunities in transportation, aerospace & defense, industrial, and sporting goods industries. The market is expected to grow at a CAGR of 6.9% from 2015 to 2020 (cf. Figure 2). The major driver for the growth of this market is growing demand for thermoplastic composites in different end use industries due to increasing requirement of lightweight, recyclable, and environment friendly products. In this market, the major reinforcements are glass and carbon fibers, whereas polyamide, polypropylene, polyetherimide, polyphenylene sulfide and polyetheretherketone are used as resins. [3]

Europe is expected to remain the largest market for CFRTPs due to growing demand for high performance, lightweight, and environmentally sustainable thermoplastic composites in different end use industries. However, North America is expected to witness the highest growth rate during the forecast period due to an expected increase in the production of advanced aircraft models, growing vehicle production and an increasing focus on lightweight composite materials. [3]
3. Approach and challenges of using thermoplastic resin

The usual process chain of CFRTP consists of five process steps: Unwinding the fibre, spreading of the fibre, manufacturing of a semi-finished textile fabric, impregnation and consolidation under high pressure. Melted thermoplastic polymers used as matrix have usually viscosities in the order of 100 to 1,000 Pa·s and high melting temperatures (>200 °C) because of their long molecular chains. Therefore, a high pressure and relatively long consolidation time is necessary to remove voids, get a good impregnation of the reinforcing textile, disperse potentially included fillers in the matrix or achieve complex geometry shapes. Furthermore, the usage of the high pressures limits the maximum part thickness, needs expensive temperature resistant tooling and introduces high thermal stresses which degrade the material properties of the polymer and results in low fatigue performance.

With the technology developed in our ongoing project it is possible to shorten the process chain and to use lower pressure in the final consolidation step. By introducing the reinforcing textile into the matrix prior to polymerization the high viscosity of the thermoplastic does not impede the full impregnation of the fibres. Subsequent to the incorporation of the mixture comprising monomers, activators and initiators the impregnated textile is consolidated under low temperature and low pressure. The most common used in-situ polymerization system in automotive applications is the anionic polymerization of ε-Caprolactam to polyamide-6 (PA6). Because of the fast process speed and the low viscosity the ε-Caprolactam process can be easily adapter to existing resin transfer molding facilities. The improved process chain with in-situ polymerization is shown in Figure 3.

Figure 2. Estimated Market Value of carbon fibre reinforced thermoplastics [3]
3. Conclusion and outlook

Via the use of in-situ polymerization for the production of CFRTPs the deficiency of the high temperature and the high pressure during consolidation can be solved. In this ongoing project a new unit is developed, which combines several steps of the conventional production process. The main advantage is the small size and thus its applicability in small research facilities. It is possible to produce small size composite parts without much offcuts, which makes the process very interesting for small and medium sized companies as well as academic research in university faculties.

The advantages of the developed system can be concluded to:

- In-situ polymerization overcomes many of the challenges using thermoplastic matrix systems
- New system is suitable to produce intermediate parts in short cycle times and good quality
- Existing resin transfer moulding facilities can be adapted
- High quality surfaces and lower thermal stress on the final parts
- Lesser limitations on the thickness of the final part

However, some challenges have to be overcome to reach mass production:

- Compatibility of caprolactam-system and carbon fibre sizing has to be tested
- Best suitable caprolactam-system (monomer, activator and catalyst) has to be found
- The precise dosing equipment and enclosure of the injection system is being developed

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