MICROWAVE CURING OF RTM PRODUCED POLYMER MATRIX COMPOSITES

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Abstract
The aim of this study was to reduce the curing time for glass and carbon fiber epoxy composites significantly. The curing time was reduced to only about one hour by the combination of microwave heating and resin transfer molding compared to at least 6 hours for conventional curing of the same material. No statistically significant differences between flexural strength and flexural modulus of microwave cured samples and conventionally cured samples were obtained. Microwave curing shows a high potential to improve efficiency of fiber composite processing without negatively affecting the mechanical properties.

1 Introduction
High specific strength, high specific modulus and customizable properties offer fiber-reinforced composites a high innovation potential for nearly all industrial fields. Up to the present times, applications are often limited by the relatively high manufacturing costs due to long cycle times and a low degree of automatization. The cycle time of the composites manufacturing process is mainly determined by the curing process of the matrix forming resin. This curing process can be accelerated by microwave processing.
Microwave heating of materials is mainly based on two mechanisms. In electroconductive materials, charge carriers get accelerated due to the electromagnetic field. This results in ohmic losses. In non-electroconductive materials microwave heating is based on dielectric polarization. Depending on the material and on the frequency range 4 different types of polarization mechanism have to be distinguished: Electronic polarization, ionic polarization, orientation polarization and interfacial polarization. The heating of polymeric matrix composites is mainly based on the orientation polarization. In contrast to conventional thermal heating, the microwave radiation generates the heating within the polymeric matrix [1,2].
Several studies demonstrated the suitability of polymer [2-6] and composite [7-13] curing by microwave radiation. Higher heating rates, a selective and volumetric heating as well as reduced energy consumption were achieved in contrast to conventional curing methods. Besides, several studies [10,13] describe an improved fiber matrix interfacial bonding due to microwave processing. Higher heating rates, due to the microwave processing, result in a lower minimum of the viscosity of the epoxy resin-hardener-mixtures [7,13]. These lower

viscosity values can promote a better fiber wetting and therefore an improved fiber-matrix interfacial strength [13].
The combination of advanced and more automated manufacturing techniques i.e. resin transfer molding (RTM) with microwave processing seems consequent. This was examined only by Papagyris et al. [13] for carbon fiber reinforced composites. In this study [13] the process time was reduced from 3 h for conventional curing to 1.5 h for microwave processing. However, the comparability of the mechanical properties between microwave and conventional cured composites was limited by different fiber volume fractions (27 % for microwave and 33 % conventional cured samples). For glass fiber reinforced epoxy composites the combination of microwave processing and resin transfer molding is not explored yet.
The aim of this study was to combine microwave curing and resin transfer molding for glass and carbon fiber epoxy composites to demonstrate the potential of this technique for industrial applications. We want to demonstrate that a significant reduction in processing time is possible without worsening the mechanical properties. Therefore, samples were produced by incorporating microwave curing into the RTM-process. Flexural strength and flexural modulus of the achieved samples were measure and compared with those of conventional cured samples.

2 Materials and testing methods

2.1 Materials

For the glass fiber reinforced composites bidirectional roving mesh (P-D Glasseiden GmbH, Oschatz, Germany) with a specific weight of 500 g/m² was used as reinforcement. The resin system used as the matrix material was L 20/EPH 943 (R&G Faserverbundwerkstoffe GmbH, Waldenbruch, Germany). The carbon fiber reinforced composites were produced of bidirectional roving mesh (Lange+Ritter GmbH, Gerlingen, Germany) with a specific weight of 380 g/m² and the resin system CR81/CH81-6 (Sika AG, Baar, Switzerland).

2.2 Sample preparation

In this study, the RTM-technique was chosen for sample preparation. The two-part mold was made of Ebalta ebaboard LX (Ebalta Kunstoff GmbH, Rothenburg ob der Tauber, Germany) and/or aluminum (see table 1). The cavity (size: 210 mm × 125 mm × 4 mm) was arranged in the lower part. In the upper element, resin supply and riser were included. The self-made injection system consists of a pressure chamber with one valve connected to a manometer to adjust the injection pressure.

	Glass fiber epoxy composites		**Carbon fiber epoxy composites**	
	Conventional	**Microwave**	**Conventional**	**Microwave**
Upper element	Ebalta ebaboard LX	Ebalta ebaboard LX	Aluminum	Ebalta ebaboard LX
Lower element	Ebalta ebaboard LX	Ebalta ebaboard LX	Aluminum	Aluminum

Table 1. Used mold materials for the preparation of conventionally and microwave cured glass and carbon fiber epoxy composites.

2.2.1 Conventional curing

7 layers of glass fibers or 10 layers of carbon fibers were placed in the cavity. Afterwards the mold was fixed. In the next step, the homogenized and degassed resin was injected under a pressure overload of 1 bar. In the case of the carbon fiber epoxy composites the mold was evacuated before the injection of the resin. After completion of the injection process, the

curing of glass fiber epoxy composites was done for 6 h at 100°C and of the carbon fiber epoxy composites for 12 h at 80°C.

2.2.2 Microwave curing

Microwave curing was done in a microwave oven MKO 30×0.85 kW (Fricke und Mallah GmbH, Peine, Germany). The microwave oven is illustrated in figure 1a. This system works with 30 magnetrons (excitation frequency 2.45 GHz), respectively 15 in the bottom and top of the microwave chamber. Each magnetron can be controlled independently. The maximum power output is about 25.5 kW. For the curing of the glass fiber epoxy resin composites a microwave configuration with a power output of 1.87 kW (22 × 0.085 kW) was used. For the curing of the carbon fiber epoxy resin composites only the magnetrons in the top of the microwave chamber were switched on. The power output of this setup was 3.825 kW (15 × 0.255 kW).

The bottom of the microwave chamber (size: 1400 mm × 1700 mm × 650 mm) can be moved vertically by six hydraulic cylinders type Enerpac RCS 502 (Enerpac, Milwaukee, USA) to apply the pressured needed for locking the RTM-mold. Each cylinder can apply a maximum force of 435 kN. The RTM-mold was fixed in the middle of the microwave chamber with a clamp. The experimental setup is illustrated in figure 1b. Filling of the RTM-mold was done identically to the conventional method.

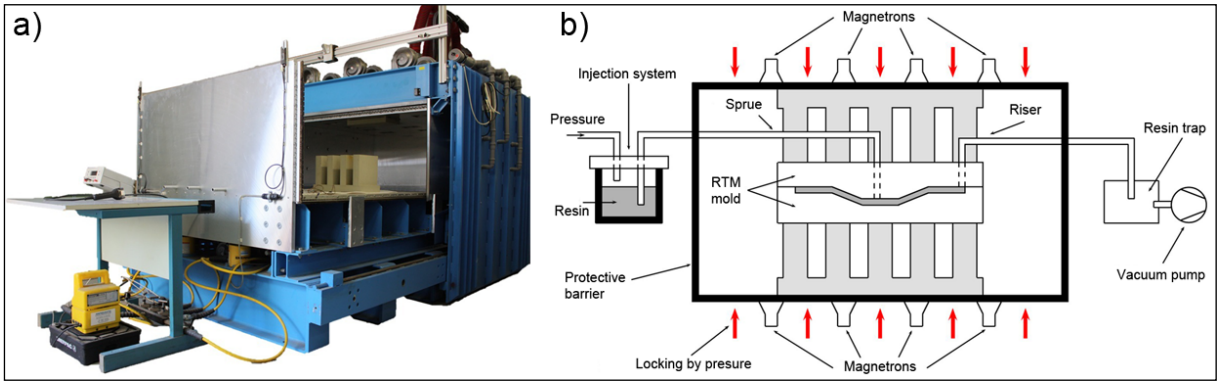

Figure 1. a) Microwave oven MKO 30×0.85 kW. b) Schematic illustration of the experimental setup for microwave curing of glass and carbon fiber epoxy composites produced by RTM.

Temperature control during the curing process was realized by using two thermo-optical sensors (Fiso Technologies Inc., Québec, Canada). A pulsed microwave heating cycle was used. First the temperature was increased to 80°C interrupted by two hold points at 40°C and 60°C. Afterwards the temperature was attuned to 75-80°C. Therefore the microwave was switched on when the temperature drops below 75°C and switched off when the temperature reached 80°C. After 1 h and 15 min, the microwave processing was stopped. The samples were immediately demolded and temperature distributions of the cured samples were measured by using an infrared camera type VarioCam head (Jenoptik GmbH, Jena, Germany).

2.3 Testing for mechanical and physical properties

Specimens for the flexural testing were cutted from the conventional and microwave cured panels according to DIN EN ISO 2818 [14]. The panel boundaries and the areas around the sprue were not used for fabrication of test specimen.

Flexural strength (σ_{fM}) and flexural modulus of elasticity (E_f) were determined according to DIN EN ISO 14125 [15]. Rectangular specimens with dimensions of 80 mm × 15 mm × 4 mm were tested using a Zwick Z020 universal testing machine (Zwick GmbH & Co. KG, Ulm, Germany) equipped with a 3-point-bending jig and a 20 kN load cell. A crosshead-speed of 2 mm/min and a span width of 64 mm were used. For each configuration 16 samples from respectively 2 panels were measured.

The measured mechanical properties were analyzed for statistical significant differences between the two ways of curing on IBM SPSS software release 19.0.0 (IBM, Armonk, USA) by a t-test ($p<0.05$).
Differential scanning calorimetry (DSC) was performed to investigate the degree of curing. Cured samples (m~10 mg) were heated in nitrogen atmosphere from 20°C to 250°C with a heating rate of 10 K/min by using a Netzsch DSC 204 F1 (Netzsch Gerätebau GmbH, Selb, Germany). From each panel three samples were tested.

3. Results

We observed an inhomogeneous heating resulting in an inhomogeneous temperature distribution during the curing of the composite samples. This is illustrated in figure 2 (left). The temperature distribution shows a previously uncured glass fiber epoxy panel heated for 75 s by microwave radiation. The inhomogeneous temperature distribution is obvious. After cooling the sample to about 80°C the sample was reheated for 70 s by microwave radiation. The resulting temperature distribution (see figure 2 (right)) illustrates that former warmer areas were less heated compared to former colder areas. In order to achieve a fast and homogenous curing of the sample an interrupted microwave power input was chosen.
By applying the described heating cycle it was possible to demold the samples after 1 h and 15 min. During the 1 h and 15 min of processing time, the microwave was switched on only for about 11 minutes.

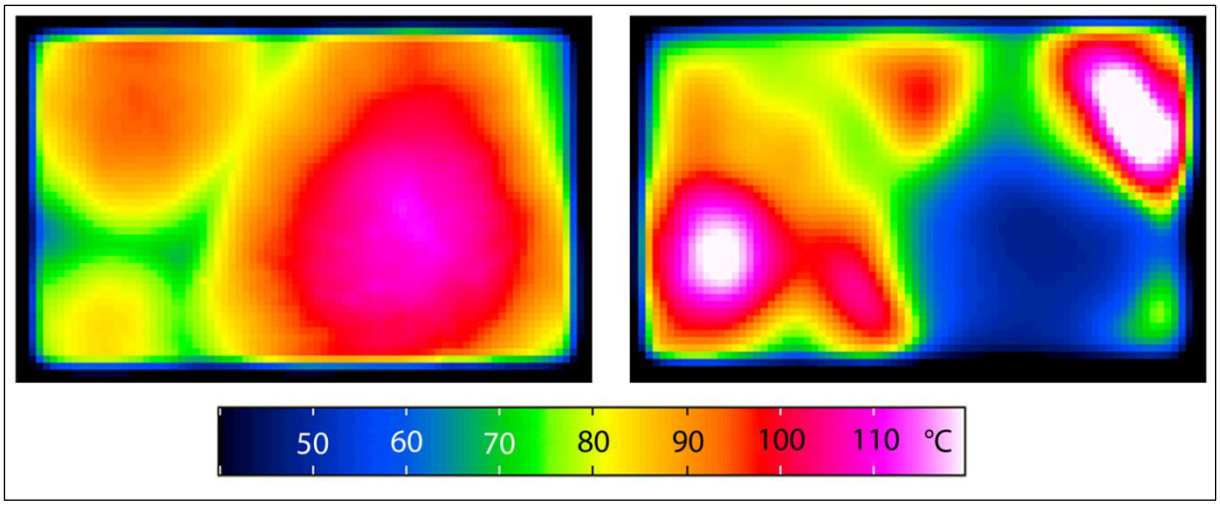

Figure 2. Left: Temperature distribution across a former uncured glass fiber epoxy sample after microwave heating for 75 s. Right: Temperature distribution of the same sample cooled to approximately 80°C and heated for 70 s afterwards [16].

The mechanical properties of microwave cured glass and carbon fiber epoxy composites and their conventional cured equivalents were compared (see figure 3). The results of the flexural test are summarized in table 2.
The flexural strength of microwave cured glass fiber epoxy composites was 417.2 ± 35.0 MPa. Thus it is slightly higher than the flexural strength of the conventionally cured samples (406.4 ± 28.8 MPa). A similar result was observed for the flexural modulus of elasticity; microwave cured composites exhibit 10.1± 0.7 GPa and conventionally cured counterparts 10.0 ± 0.8 GPa. Microwave cured carbon fiber epoxy composites exhibits a flexural strength of 664.1 ± 39.5 MPa, for the conventional cured samples a slightly increased value of 672.8 ± 49.1 MPa was measured. The flexural modulus was 43.2 ± 5.4 and 42.8 ± 1.9 GPa for microwave and conventionally cured samples, respectively.

However, statistically significant differences between the observed mechanical properties of the microwave and conventionally cured samples were not found.

	Glass fiber epoxy composites		Carbon fiber epoxy composites	
	Conventional	**Microwave**	**Conventional**	**Microwave**
Flexural strength [MPa]	406.4 ± 28.8	417.2 ± 35.0	672.8 ± 49.1	664.1 ± 39.5
Flexural modulus [GPa]	10.0 ± 0.8	10.1 ± 0.7	42.8 ± 1.9	43.2 ± 5.4

Table 2. Mean value and standard deviation for flexural strength and flexural modulus of conventionally and microwave cured glass and carbon fiber epoxy composites [16].

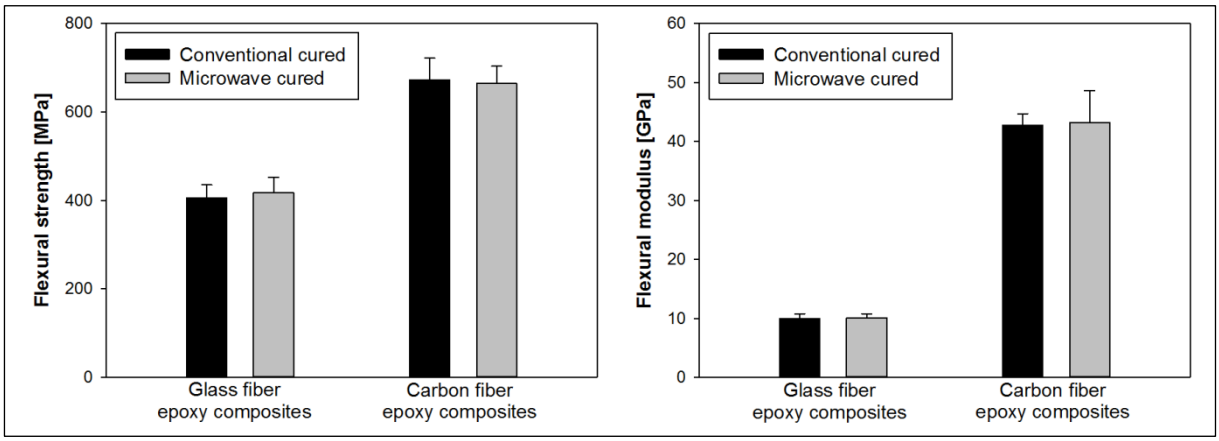

Figure 3. Flexural strength and flexural modulus of conventionally and microwave cured glass and carbon glass fiber epoxy composites [16].

For none of the investigated samples an exothermic reaction was observed during the DSC heating process. Therefore it seems that microwave cured panels as well as conventional cured panels were cured fully.

4. Discussion

The application of fiber reinforced composites in large-volume production is often limited due to their long cycle time. The cycle time is mainly determined by the curing time of the resin-hardener-mixture, this corresponds with the time between filling of the mold and demolding of the construction element. One possibility to reduce the cycle time is to combine an automated production technique, i.e. the RTM-process, with a microwave process for the curing of the resin-hardener-mixture. We showed that microwave processing allows reducing the cycle time from at least 6 h for conventionally curing to almost 1 h. This reduction of cycle time is due to higher heating rates and a selective heating permitted by the microwave processing. In general, a further reduction of the curing time is imaginable. In this study the maximal temperature was limited by the mold material. Temperatures higher than 90 °C resulted in a damage of the mold parts made of Ebalta ebaboard LX. Therefore, a next step for a further improvement of the presented manufacturing technique is the search for a more suitable mold material. An optimized mold material should combine transparency for microwave radiation, temperature stability and good processability.

In this study, the electromagnetic field was not homogenous. This results in different heating rates inside the panel. However, the dielectric loss is higher for uncured resin-hardener-mixture than for cross-linked equivalents [13]. Figure 2 (left) shows the temperature distribution across a former uncured glass fiber epoxy sample heated for 75 s. The temperature distribution inside the panel is not uniform. Most likely, the degree of curing

resulting from this heating cycle was higher in warmer areas. After cooling to approximately 80°C the same sample was heated again for 70 s. From figure 2 (right) is obvious that areas with a lower degree of curing resulting from the first heating interval were heated faster than areas with a higher degree of curing due to the change in dielectric properties. Therefore, a homogenous hardening can be achieved by using an interrupted power input during microwave curing process as described above. This result agrees with the study of Tanrattanakul and Jaroendee [10].

Besides the reduction of the processing time the mechanical properties of the microwave cured composites were of major interest in this study. The statistical analysis showed no significant differences between the flexural strength and the flexural modulus of microwave and conventionally cured glass or carbon fiber epoxy composites. Therefore, we found no evidence for an improved fiber matrix interfacial bonding due to the microwave processing as described elsewhere [10,13].

5. Conclusion

The incorporation of microwave heating into the RTM process leads to a significant reduction of the cycle time during the production of glass and carbon fiber epoxy composites. Panels of 210 mm × 125 mm × 4 mm in size were homogenously and reproducible cured by the usage of a well-adapted appropriated design of the microwave configuration and an optimized heating program. Mechanical properties of microwave cured samples were compared with conventionally cured samples. The mean values for flexural strength and flexural modulus of elasticity were found to be not statistically significantly different between microwave and conventionally cured samples. Thus the combination of microwave curing and RTM process offers a serious potential to improve the efficiency in industrial manufacturing of glass and carbon fiber reinforced composites.

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