Development of glass fibre reinforced composites using microwave heating technology

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Abstract. Fibre reinforced composites are differentiated by the used matrix material (thermoplastic versus duroplastic matrix) and the level of impregnation. Thermoplastic matrix systems get more important due to their suitability for mass production, their good shapeability and their high impact resistance. A challenge in the processing of these materials is the reduction of the melt flow paths of the thermoplastic matrix. The viscosity of molten thermoplastic material is distinctly higher than the viscosity of duroplastic material. An approach to reduce the flow paths of the thermoplastic melt is given by a commingling process. Composites made from commingling hybrid yarns consist of thermoplastic and reinforcing fibres. Fabrics made from these hybrid yarns are heated and consolidated by the use of heat pressing to form so called organic sheets. An innovative heating system is given by microwaves. The advantage of microwave heating is the volumetric heating of the material, where the energy of the electromagnetic radiation is converted into thermal energy inside the material. In this research project microwave active hybrid yarns are produced and examined at the Institute for Textile Technology of RWTH Aachen University (ITA). The industrial research partner Fricke und Mallah Microwave Technology GmbH, Peine, Germany develops an innovative pressing systems based on a microwave heating system. By implementing the designed microwave heating technology into an existing heat pressing process, FRTCs are being manufactured from glass and nanomodified polypropylene fibre woven fabrics. In this paper the composites are investigated for their mechanical and optical properties.

1. Introduction

In the context of the “Micropress” project, which is supported by the central innovation programme (ZIM) of the federal ministry for economic affairs and energy, the manufacturing of FRTCs with an innovative heating method is being investigated by the ITA. This project is based on researches dealing with nanomodification and commingling of hybrid yarns to achieve reduced processing times in the final manufacturing process [1]. The reduction of manufacturing cycle times is essential for targeted series-production of fibre reinforced plastics (FRP) in general. Therefore, innovative materials and material combinations as well as manufacturing methods are being developed. The considered research is focused on the application of a heat pressing process modified with microwave technology. A review of the crucial aspects with focus on the FRTC production and analysis of the research is given in this paper.
FRTCs display several advantages compared to thermosetting composites, e.g. a potential for shorter manufacturing cycle times, a higher shelf life and an improved recyclability [2] [3]. By commingling thermoplastic and reinforcing material on fibre basis, even high-viscosity matrices become processable and suitable for mass production. This is enabled by short flow paths of the thermoplastic matrix material, which are limited to a maximum of few millimetres [2] [3]. The chosen hybrid yarn type for this research is a multifilament hybrid yarn consisting of polypropylene (PP) and glass filaments (GF). The blending of the two components is achieved by air jet mixing (commingling) [4]. Heat pressing is also suitable for series-production due to a high degree of automation, short cycle times and a high reproducibility. Since the low thermal conductivity of plastics, the applied heating methods cause long heating times increasing the overall manufacturing cycle time [3].

Despite their widespread proliferation, infrared and hot plate heating methods exhibit particular disadvantage like a decreased controllability or increased space requirements [3] [4]. In addition to that, the surfaces of the applied moulding tools are supposed to be coated with appropriate materials to avoid adhesion of the matrix material on the moulding tool [4] [5]. In contrast to the application of heat through conduction, the microwave based heating method is dependent on activatable additives in the thermoplastic material.

The main advantage of this method is a volumetric heat application inside the moulding part reducing the heating time. The microwave technology exhibits further advantages like high energy efficiency and contactless energy application. Applicable additives are carbon based structures, silicon carbide, titanium dioxide and metal flakes. Beyond that, nanoscale particles like carbon nanotubes (CNT) with a large surface area have been found to be very effective at a suitable distribution inside the plastic [6] [7].

2. Experimentation
In the following chapters, the experiments which are carried out along the manufacturing process of the FRTCs are described. These experiments are focused on the consolidation process by microwave technology.

2.1. Applied materials
The thermoplastic component of the FRTC is a PP, which is modified with nanoscale CNTs. The mass fraction of CNTs is about 1%. The PP is available in the form of a multifilament yarn with a linear density of approximately 396.53 tex and a mean density of about 0.9106 g/cm³. The multifilament yarns have been manufactured at the ITA to achieve low agglomeration of CNTs improving the heat development in heat pressing. The reinforcing fibres are glass filaments, which are available in the form of a multifilament yarn with a linear density of ca. 597.2 tex and a mean density of about 2.617 g/cm³. The multifilament yarn is a product of PPG Industries, Inc., Pittsburgh, Pennsylvania, USA with the type designation TUFRov 4599. Moreover, the fibre is provided with a sizing to improve bonding with the PP matrix. Both fibres are mixed by commingling to hybrid yarns that consist of 40 wt% PP and 60 wt% glass fibres with in total 994 tex. In a second step this hybrid yarns are processed to woven fabrics.

2.2. Microwave heat pressing
Unidirectional fabrics from chosen hybrid yarns are processed to FRTCs by microwave heat pressing. The microwave technology is implemented into a press by Fricke und Mallah Microwave Technology GmbH, Peine, Germany. The installed tool can be used to manufacture FRTCs with a maximum size of 400 x 400 mm² and the power can be controlled from 500 W to a maximum of 5000 W. The manufactured FRTCs consist of two or four layers of unidirectional fabrics. These preforms are heated and consolidated by the microwave heat pressing process and optically and mechanically examined.
subsequent to manufacturing. This includes light microscopic images of cross-sectional areas, tensile and flexural tests.

3. Results of microwave testing
The results of the executed experiments are presented and discussed in the following chapters.

3.1. Microwave heating experiments
The measured temperatures in these experiments describe rather the surface temperature of the quartz glass load than the actual temperature of the specimen. In this context, the temperature difference to the starting temperature is shown in the diagram (Σφάλμα! Το αρχείο προέλευσης της αναφοράς δεν βρέθηκε.). In addition to that, several thermal images recorded along the heating experiment are shown as well to clarify the heat development. The experimental setup in the microwave oven is schematically illustrated in Figure 1.

![Figure 1. Temperature differences of a hybrid yarn specimen measured by thermal imaging of a heating experiment with a power of 325 W](image)

The hybrid yarn specimen of this experiment is irradiated by microwaves with a power of 325 W. The two graphs in the diagram show increasing temperatures with advancing irradiation time. The maximum temperature difference $\Delta T_{\text{max}}$ indicates a high temporal temperature gradient at first which decreases after ~ 10 s. Moreover, the temperature difference rises steadily till 100 s and sinks afterwards to show a second rise at about 120 s irradiation time. Also, the mean temperature difference $\Delta T_{\text{mean}}$ shows a positive temporal temperature gradient which increases at about 120 s. It can be concluded from the temperature differences and the thermal images that the specimen is heated locally.
at first. The heated area is shifted along the specimen with advancing time, before a volumetric heating begins. This volumetric heating is determined by the increasing temporal temperature gradient of the mean temperature difference. Furthermore, the reached temperature difference of about 165 K clearly indicates that the PP filaments have been heated sufficiently to gain the melt temperature of 160 °C to 165 °C. The actual temperatures are presumably higher, because the specimen shows signs of thermal degradation after the heating experiment.

3.2. Heat pressing of specimens

To perform a start-up of the innovative processing technology, specimens with various sizes of surface area and number of layers are produced by a stepwise adjustment of applied pressure and irradiation time. The interaction of the microwave radiation with the preform is based on a complex relation between the electromagnetic field, the filaments and the nanoparticles. As these are all inhomogenously distributed, the preforms are heated carefully reducing the risk of hot spots which can lead to thermal degradation of the matrix. The performed tests which lead to a sufficient heating of the specimens are shown in Table 1.

Table 1. Microwave heat pressing tests.

| Test notation | Surface area A [mm²] | Applied pressure p [bar] | Power P [% of maximum] | Number of layers n [-] | Irradiation time tI [min] | Starting tool temperature T₀ [°C] |
|---------------|----------------------|--------------------------|------------------------|-------------------------|---------------------------|-------------------------------|
| MW1           | 200 x 180            | 10.9                     | 70                     | 2                       | 15                        | 25                            |
| MW2           | 200 x 180            | 8.2                      | 60                     | 2                       | 6                         | 145                           |
| MW3           | 300 x 190            | 5.5                      | 40                     | 4                       | 5                         | 150                           |
| MW4           | 330 x 200            | 5.9                      | 35 to 45               | 4                       | 42                        | 50 to 160                     |
| MW5           | 120 x 80             | 8.2                      | 45                     | 4                       | 10                        | 25                            |
| MW6           | 300 x 130            | 6.0                      | 45                     | 4                       | 8                         | 130                           |
| MW7           | 300 x 130            | 6.0                      | 45                     | 4                       | 9                         | 130                           |
| MW8           | 300 x 130            | 6.0                      | 45                     | 4                       | 10                        | 130                           |

A certain tool temperature has to be reached before a sufficient amount of energy can be transformed to melt the thermoplastic filaments. This is conditioned by the surface area of the tools causing a high heat flow away from the specimen by heat conduction. The produced FRTCs of tests MW1, MW2 and MW3 exhibit degraded matrix material due to the development of hot spots. Moreover, the applied pressure forces the matrix to flow out of the specimens. A further adjustment of specimen size and applied pressure along with a reduced power leads to insufficient heating of the specimen at the beginning of test MW4. The power is increased stepwise to 45 % also causing an increasing tool temperature. The minimal power level providing sufficient heating is found at 45 % of the maximum power. According to this result, three proper FRTC specimens are produced (MW6 to MW8). Test MW5 serves as tool heating intermediate step.

3.2.1. Optical analysis

The manufactured FRTCs are optically examined by light microscopic images of cross sectional areas. This delivers insight into the specimens and allows an evaluation of the impregnation and consolidation quality of the microwave heat pressing. Two images of cross-sectional areas of specimen from tests MW6 and MW7 are shown in Figure 2. In addition to that, the dotted areas are
zoomed in for enhanced visibility. The images show that the impregnation and consolidation quality is partially insufficient. There are sections with imperfect impregnation. These sections are located inside of the glass rovings. Moreover, there are air bubbles and voids inside of the specimens. The lower thickness of the specimen from test MW7 indicates that inhomogeneous material distribution also leads to a varying pressure distribution inside the preform. The width of the voids is significantly higher compared to the specimen from MW6. It can be determined that suitable process parameters have to be found in future research to utilize the advantages of microwave heat pressing. These include the applied pressure, power level, irradiation time as well as applicable materials and material combinations.

![Figure 2](image_url)

**Figure 2.** Light microscopic cross-sectional images of probes from MW6 and MW7

#### 3.2.2. Mechanical analysis

The mechanical analysis of the manufactured FRTCs provides important characteristic values like breaking strength and Young’s modulus. Probes for the mechanical tests are cut out of the specimens of MW6 and MW7 because of their high impregnation and consolidation quality. Furthermore, the mechanical tests are performed according to DIN EN ISO 527-5 (tensile test) and DIN EN ISO 14125 (flexural test). The volume fraction of the glass filaments is at 34 %. The gathered data of the executed tests is summarized in Table 2.

**Table 2.** Characteristic values of the tensile and flexural tests.

| Characteristic value (mean value ± deviation) | Breaking strength $\sigma$ [MPa] | Strain at breaking strength $\varepsilon$ [%] | Elasticity modulus $E$ [GPa] |
|---------------------------------------------|----------------------------------|------------------------------------------|-------------------------------|
| Tensile test                                | 190.64 ± 35.28                  | 1.61 ± 0.25                              | 14.19 ± 1.43                 |
| Flexural test                               | 109.70 ± 14.60                  | 2.42 ± 0.44                              | 6.07 ± 2.77                  |
These results are at the same magnitude as conventionally produced samples of unidirectional PP/GF-composites. This leads to the assumption, that the processing of FRTCs with a microwave heated press achieves at least comparable mechanical properties. However, these results have to be depend in future research to evaluate the improvement of the microwave heat pressing process by adjusted processing parameters or improved microwave active materials. This could lead to a significant improvement of the processing times and impregnation quality compared to the conventional process.

4. Results
A valid temperature measurement of the material is crucial for predicting suitable microwave heat pressing parameters. The applied thermal imaging could be replaced by a fibre based temperature sensor providing precise data. Summarizing the presented results, it can be said that the manufacturing of FRTCs by microwave heat pressing is practicable. To exploit the complete advantages provided by the use of microwave technology, it is inevitable to consider the whole manufacturing process starting with the composition of applicable materials and ending with the heat pressing process parameters. The required cycle times for the start-up of the microwave heat pressing are found to be around 10 minutes with a pre-heated tool. These results show that the potential offered by microwave technology is not utilized to its full extent as can be seen according to the research in [6]. A temperature rise of more than 80 °C is achieved in 20 s by the use of polymers filled with CNTs at a mass fraction of 1 %. Therefore, it is advised to carry out further, extensive research in consecutive projects. This can enhance the observed impregnation and consolidation quality as well as the required manufacturing cycle times significantly. The theoretical and practical comprehension of transformation of electromagnetic energy by polymer and filler materials along with an aligned processing technology could clearly result in an able method for series-production of FRTCs.

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