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Influence of parameters of the production process on the material quality of unidirectionally reinforced prepregs

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ABSTRACT

Purpose: A prepreg production device in laboratory scale is used to develop the production process of unidirectionally reinforced prepregs.

Design/methodology/approach: The aim of the prepreg production device is to impregnate different types of reinforcement fibers with an arbitrarily selectable thermoset matrix system that completely satisfies the requirements for autoclave processing. As the prepreg production device is designed and built up modularly every module corresponds one step in the process.

Findings: To identify the parameters of the production process and investigate its sensitivity on the material quality of both the prepreg as an uncured semi-finished product and the composite as the cured material experimental investigations regarding the resin flow, fiber volume content, mass per unit area and void content are carried out. Overall four material combinations have been investigated, where in each case the selected impregnation temperature and the width of the impregnation gap has been reproducibly varied in selected steps.

Research limitations/implications: The experimental characterization of the prepregs and of the composite material is carried out according to German standards.

Originality/value: Used parameters clearly affect the material properties, so that a proper impregnation and curing process can be achieved by optimizing the parameter to desired values.

Keywords: Autoclave processing; Fiber volume content; Hot-melt-impregnation; Prepreg; resin flow; Unidirectional layer; Void content

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MANUFACTURING AND PROCESSING

1. Introduction

New materials are a basis for technical innovation. To define any new materials' application spectrum, it is necessary to know the individual advantages and especially the structural mechanical properties of these materials. Since fiber reinforced plastics offer great lightweight potential due to its outstanding high specific stiffness and high specific strength regarding its density lately industrial applications significantly increase, among in aircrafts, wind power plants or electric vehicles. That is the reason why a prepreg-application ("*pre*-im*preg* nated fiber") for unidirectional prepregs with an independently selectable thermoset matrix system has been designed and built up. The prepreg production device offers the possibility to produce prepregs as two-dimensional semi-finished products in amounts that are suitable for laboratory scale, but would not be cost-effective and payable on an industrial production plant. The produced unidirectional layers are used for the layup of test panels that are cured in autoclave processing in order to obtain test panels for specimens [1,2].

2. Motivation

The motivation to design and build up the afore mentioned device results from the lack of independently selectable combinations regarding the single components, namely reinforcement fiber and matrix system, as a prepreg. There are prepreg systems with common reinforcement fibers, like carbon, glass or aramid fiber with different types of resin, that cannot be acquired in an independently selectable combination, but in established ones. In order to experimentally investigate the influence of different kinds of reinforcement fibers on structural mechanical properties the presence of one and the same matrix system is an essential requirement. If this requirement is not fulfilled it is not possible to explicitly identify the influence of the kind of fiber reinforcement on the structural mechanical properties of the composite material. In order to obtain the required combination of fiber and matrix for experimental investigations it would also be possible to individually order the material. Due to the cost-effective production process of a prepreg plant this would oblige to buy one whole charge. This amount, however, is suited for industrial production scales but exceeds laboratory scales by far.

The German standard DIN 65378 [3] specifies the production of unidirectionally reinforced single layers by the filament winding technique with very low viscosity resins. However, the process suffers high efforts regarding costs and time. Additionally the obtained material quality often is poor, because necessary manual steps cause derivations and reduce the reproducibility.

There is also the possibility to buy fabric reinforced prepregs or manually impregnated woven fabrics. But as a structural mechanical characterization of composite materials is based on unidirectional layers and a manual process always suffers deviations, it is at the same time technically and economically reasonable to design and build up the afore described device [4].

3. Technical methods for fiber impregnation

There are several production processes for fiber reinforced plastics. The one that provides at the same time the highest structural mechanical properties for the resulting composite material and the highest reproducibility is autoclave processing. In autoclave processing the material is cured under a defined temperature and pressure cycle during the process time. For autoclave processing prepregs as special semi-finished products are typically used. The viscosity of the resin during impregnation process is a significant parameter. Common prepreg resins are high-viscous at room temperature in order to provide a nearly dry handling of the prepregs while doing the layup of the later composite. This nearly dry state of the prepregs at room-temperature is called the B-stage [2]. It is a determined state of the thermoset matrix system that can only be maintained by storing the prepreg at a temperature of -18 °C and defreezing it for the duration of the layup process. The impregnation of the reinforcement fibers is also called the consolidation process. The aim of this process is to properly impregnate every monofilament of a single roving with resin. The two most popular methods are described in the following [1,5].

3.1. Solvent-impregnation

The solvent-impregnation method uses solvent to decrease the viscosity of the resin. For consolidation the straightened fibers are guided through the solvent diluted resin. Nip rollers assure the distribution of the solvent diluted resin around the monofilaments. Optimizing the amount of remaining resin finally yields the desired fiber volume content in the later cured composite material. A heated drying chamber removes the solvent nearly completely in order to obtain the semi-finished product. To properly store the prepreg it gets covered on bottom and top side by foil and is wound up a coil of sufficient diameter [1, 5, 6].

3.2. Hot-melt-impregnation

For this impregnation process a foil covered with the resin is necessary. The resin covered foil is deposited on the tape on both sides. A heated roller mechanism melts the resin to lower its viscosity and applies pressure on the foils to impregnate the fibers. Cooling down the prepreg to room temperature is the final step of the process, before winding up the prepreg in order to store it [1, 2, 5, 6].

3.3. Selected impregnation process

In the prepreg device a combination of both formerly described methods is used. An immersion bath similar to those used in filament winding technology, is equipped with a heating unit and built up in an isolated case. A controlled heating up process of the resin to selected temperatures decreases the viscosity of the resin in order to properly consolidate the fibers with the resin.

4. Configuration of the machine and preparation for the production

The prepreg production device is designed and built-up in a modular way. Each module corresponds to one single step of the production process. The aim is to render the device expandable for optional steps in the production process for special requirements. The device provides a maximal amount of 30 coils on which rovings are wound up. Each coil's capacity is approx. 55 m of rovings out of glass fibers with 2400 tex or other kind of fibers with respective linear density. Thus the machine is able to produce approx. 50 m of UD prepreg with a maximum width of approx. 200 mm. Figure 1 illustrates the different modules that are described in detail in the following sections.

4.1. Unwind stand

The first step of the production process consists of two parts. They are the orientation of the single rovings into a unidirectional layer or tape and the application of a defined pre-stress. The pre-stress is necessary to properly impregnate the fibers with the resin, as exemplarily specified in the German standard DIN EN ISO 9163 [7]. A strap brake on each roving coil generates a mechanical braking torque which results in a definable pre-stress on every single roving while unwinded. In summary the force on the brake straps decreases during the production process automatically whereas the pre-stress on the fibers remains constant. As the force applied by the brake straps decreases over the steps of the process the pre-stress on the fibers can be presumed to remain constant over the process. All the pre-stressed rovings are guided through a slewable comb with 30 pins which switches the rovings in an angle of 90°. This change of direction assures the unidirectional orientation of the fibers. The angle of the comb directly affects the ratio of thickness and width of the layer. In conclusion, the unwind stand delivers a defined pre-stressed unidirectional layer of reinforcement fibers for the next step of the process.

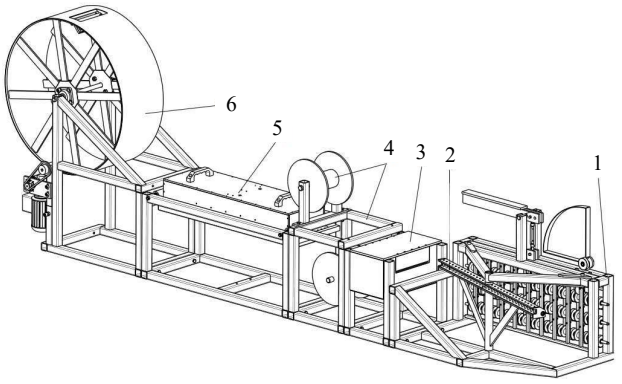

Fig. 1. Prepreg production device in laboratory scale: 1 unwind stand, 2 slewable comb, 3 impregnation module, 4 protective foil module, 5 reeling device

4.2. Impregnation module

In the first part of the second step the initially dry tapes are impregnated in the afore described method. Fig. 2 shows the cross-section of the module. The initially dry tape is guided through the reservoir containing the resin. The system of rolls assures the impregnation from bottom and top side of the tape. Guiding the impregnated tape through a selected gap between two further rolls, affects the fiber volume content to a selectable value as the gap is adjustable. The excessive resin flows back into the reservoir. In order to reproducibly adjust the gap a distance gauge is used.

To assure a constant matrix level for the entire duration of the process, the bottom part of the case is mounted on a scissor lift. The mechanism elevates the matrix container to counteract the decreasing matrix level during the process. In order to decrease the viscosity of the resin, the reservoir enclosed by an isolated case is heated up to selected temperatures lying in the range from 30 °C to 70 °C. A

constant temperature is assured by a two-position controlled heating system with heating foils inside the case.

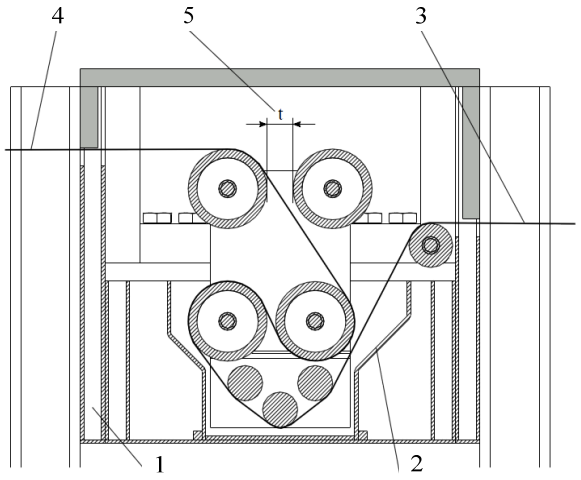

Fig. 2. Cross-section of the impregnation module: 1 isolated case, 2 matrix reservoir, 3 dry unidirectional layer of fibers, 4 uncovered prepreg, 5 variable gap for fiber volume content adjustment

4.3. Protective foil module

As a protection and in order to enable a proper handling of the tacky impregnated unidirectionally reinforced layer, it is covered by thin plastic foils out of polyethylene (PE) from bottom and top side. Therefore two coils for foil storage are mounted on the device. Each coil is equipped with a brake to slightly pre-stress the foils and to prevent wrinkles while covering. A redirection of both foils is installed, so that the prepreg gets covered immediately after the impregnation module and to protect it from contamination and damages.

4.4. Reeling device

The reeling device consists of a coil that is driven by a geared electric motor by a chain drive system. It is at the same time the prepreg storage and actuator for the production process. To prevent corrugations and ondulations of the prepreg after taking it off the coil a diameter of 800 mm is used [8].

4.5. Adjustable parameters

The device provides selected parameters that can reproducibly be varied in order to investigate its influence on the produced prepreg. In detail the parameters are:

- the pre-stress on the fibers,
- the thickness-width ratio of the ribbon,
- the temperature of the matrix system,
- the width of the gap in the impregnation module,
- the pre-stress on the cover-foils and
- the winding velocity.

This paper describes the experimental investigations of varying the width of the gap in the impregnation module. The aim is to identify its influence on the fiber volume content, define its sensitivity and optimize it to a desired value that is suitable for a proper use of the material in autoclave processing with common curing cycles. Therefore experimental investigations according to the German standards DIN EN ISO 1172 [9] and DIN EN 2332 [10] regarding the fiber volume content and the resin flow, respectively, are carried out. The procedure and its result are described in the following.

4.6. Preparation for the production of unidirectionally reinforced prepregs

Several preparations are necessary to start the continuous process. The corresponding steps are briefly described in the following sections.

4.6.1. Winding the coils

To charge the prepreg machine with the reinforcement fibers, it is necessary to reel up the rovings on the coils which have been described before. As the rovings initially are delivered as a spool without tube for internal unwinding, according to one possible delivery configuration specified in the German standard DIN 65060 [11], a rewinding process on the much smaller coils, that suit to the unwind stand (1st module), is necessary. Therefore a rewinding unit that is not part of the prepreg production device, but yet essential for a proper distraction of the roving, has been designed and built up. It realizes a cross winding, which is necessary for pre-stressed unwinding during the process without any difficulty [12].

4.6.2. Feeding the rovings through the device's modules

In order to start the continuous process with the actuator at its end, the rovings have to be fed through the device's modules. Therefore the coils are mounted in the unwind stand. The end of each roving is led through the corresponding guiding in the comb. After fixing all single rovings parallely by a clamp it is possible to feed the device through its modules by simply guiding the clamp. Finally the clamp is fixed with rovings and the protective foils on the coil of the reeling device.

4.6.3. Mixing and preheating the thermoset resin

The resin is mixed according to the ratio indicated in the respective technical data sheet of the three selected thermoset matrix systems, as listed in Table 1. After decanting the mixed resin into the reservoir of the impregnation module the preheating is started and controlled by the afore described mechanism.

5. Materials, processing and experimental investigations

In the test series, besides the impregnation temperature, the width of the gap in the impregnation module has been varied in selected steps, as further described in section 5.2. and indicated in Table 2. In order to properly impregnate a glass roving of 2400 tex the German standard DIN EN ISO 9163 [7] specifies an eye diameter of 1.4 mm to achieve a technical reasonable fiber volume content. In this case, a gapwidth of 0.38 mm and a tape-width of 120 mm corresponds to the thirty-fold cross-section area for 30 rovings, compared with the eye diameter. Regarding a reasonable range for the variation of this parameter prepregs have been produced with a gapwidth in the range from 0.30 mm to 0.55 mm in steps of 0.05 mm, as listed in Table 2. Thereby, all other parameters have been held constant. The afore mentioned variation of the impregnation temperature has been carried out by the production of another lot produced with the different selected impregnation temperature.

5.1. Materials

In detail two kinds of anorganic reinforcement fibers have been selected for the investigations. The two kinds of reinforcement fibers are namely an E-glass fiber named 076 PR 2400-1 as a direct roving [13, 14] provided by Mühlmeier GmbH & Co. KG and a basalt fiber named BR130.24000.12RAA as an assembled roving [15] provided by Incotelogy Ltd. Both rovings have the linear density of 2400 tex. Due to their relatively equal density of E-glass and basalt the cross-section of the rovings are relatively equal, too. The relevant mechanical and physical properties of the reinforcement fibers are listed in Table 3.

Further three kinds of warm-curing thermoset resins as polymeric matrix systems have been selected for the investigations. They are epoxy resins in each case. They are named Epikote [16] provided by Momentive Performance Materials Inc., CeTePox [17] provided by CTP Advanced Materials GmbH and Sicomin [18] provided by Sicomin Epoxy Systems. In each case the epoxy resins exhibit a relatively low viscosity, what makes them suitable for both the selected impregnation process, described in the previous section 3.3. and the autoclave processing, described in the following section 5.3. The relevant mechanical and physical properties of the polymeric matrix systems are listed in Table 1. Regarding the autoclave processing the relevant parameters for autoclave curing of the warm-curing, low-viscosity epoxy resins are listed in Table 4.

5.2. Material combinations and parameter variations

Overall four material combinations have been investigated. Thereby the width of the impregnation gap has been varied from 0.25 mm to 0.50 mm in five equidistant steps of 0.05 mm. In each case the selected impregnation temperatures are 30 °C and 50 °C, whereat regarding the first material combination a more detailed variation from 30 °C to 55 C in steps of 5 °C has been applied. Table 2 lists the selected material combinations of reinforcement fibers and polymeric matrix systems with the respective steps of impregnation temperature and widths of the impregnation gap.

Table 1.
Relevant mechanical and physical properties of the polymeric matrix systems

Matrix system	Type of material	Trade name/ components	Provided by	Density ρ at 20 °C	Dyn. viscosity η at 25 °C	Curing temp T_c	Literature
Epikote	Thermoset plastic (epoxy)	Epikote Resin 05128, Epikure Curing Agent 3280	Momentive Performance Materials Inc.	1.17 g/cm^3	8-12 Pa·s	120 °C	[16]
CeTePox	Thermoset plastic (epoxy)	CeTePox AM VP 382-1 A, CeTePox AM VP 382-1 B1, CeTePox AM VP 382-1 B2	CTP Advanced Materials GmbH	1.17 g/cm^3	10-12 Pa·s	110 °C	[17]
Sicomin	Thermoset plastic (epoxy)	SR 8500, KTA 21 3280	Sicomin Epoxy Systems	1.176 g/cm^3	4-5 Pa·s	110 °C	[18]

Table 2.
Material combinations and parameter variations

Reinf. fiber	Matrix system	Temp. at imp. T	Width of imp. gap
E-glass	Epikote	30 °C	0.25 mm
		35 °C	0.30 mm
		40 °C	0.35 mm
		45 °C	0.40 mm
		50 °C	0.45 mm
		55 °C	0.50 mm
E-glass	CeTePox	30 °C	0.25 mm
			0.30 mm
			0.35 mm
		50 °C	0.40 mm
			0.45 mm
			0.50 mm
E-glass	Sicomin	30 °C	0.25 mm
			0.30 mm
			0.35 mm
		50 °C	0.40 mm
			0.45 mm
			0.50 mm
Basalt	Sicomin	30 °C	0.25 mm
			0.30 mm
			0.35 mm
		50 °C	0.40 mm
			0.45 mm
			0.50 mm

5.3. Experimental characterization of the prepregs and of the composite material

Regarding the further experimental material characterization, as described in the following section 6, the layup and the lateral dimensions of the test panels as well as the layup of the vacuum bag and the autoclave pressure p have been chosen according to the German standard DIN 2332 [10]. In detail of each material combination and every parameter variation indicated in Table 2 one test panel with $[0/90]_s$ layup of the unidirectional prepreg tapes has been produced with sufficiently large lateral dimensions of approx. 100 mm x 100 mm.

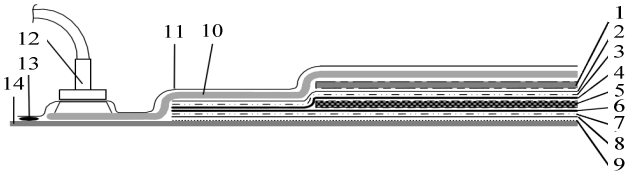

Fig. 3. Schematic cross-section of the layup for measuring the resin flow according to the German standard DIN EN ISO 2332 [10]: 1 top aluminum board, 2 peel ply, 3 additional glass fiber fabrics, 4 perforated foil, 5 prepreg layup, 6 perforated foil, 7 additional glass fiber fabrics, 8 peel ply, 9 release film, 10 bleeder, 11 vacuum bag, 12 vacuum connector, 13 sealant tape 14 bottom aluminum board.

In autoclave processing pressure and temperature is applied on an evacuated layup under a vacuum bag to cure the composite material. Among the several production processes for composites autoclave processing ensures the highest structural mechanical quality and highest degree of reproducibility at the same time. Figure 3 schematically shows the cross-section of the layup for measuring the resin flow according to the German standard DIN EN 2332 [10]. Table 4 contains the curing temperature Tc, the autoclave pressure p and the relative vacuum applied during the curing time t1 as the relevant parameters of the autoclave curing cycle for each selected polymeric matrix system according to the resin's datasheet, the German standard DIN EN 2332 [10] and [6, 19, 20], respectively.

Table 3.
Relevant mechanical and physical properties of the reinforcement fibers

Reinforcement material	Type of material	Trade name	Provided by	Density ρ	Linear density	Filament diameter	Moisture	Literature
E-glass fiber	Direct roving	076 PR 2400-01	Mühlmeier GmbH & Co. KG	2.6 g/cm^3	2400 tex	20 µm	0.008%	[13, 14]
Basalt fiber	Assembled roving	BR130.2400.12RAA	Incotelogy Ltd.	2.7 g/cm^3	2400 tex	13 µm	< 0.1 %	[15]

Table 4.
Relevant parameters of the polymeric matrix systems for autoclave curing

Matrix system	Curing temp. T_c	Autoclave pressure p	Relative vacuum	Curing time t	Literature
Epikote	120 °C	4 bar	-0.1 bar	230 min	[19]
CeTePox	110 °C	4 bar	-0.1 bar	620 min	[17]
Sicomin	110 °C	4 bar	-0.1 bar	260 min	[18]

5.4. Experimental characterization of the prepregs and of the composite material

In the following the experimental characterization of the prepregs and of the composite material are briefly described. The equations for evaluating the experimental results are indicated with their respective standard.

5.4.1. Resin flow

In order to characterize the transformation process from the uncured layup of prepregs to the resulting composite material by curing experimental investigations regarding the resin flow are carried out. The resin flow indicates the relation between flown out resin during the curing process to the resin in the resulting composite material. The resin flow is based on weighted values. Its experimental determination is carried out according to the German standard DIN 2332 [10]. Instead of the required heatable press an autoclave has been used.

A $[0/90]_s$ layup of unidirectional prepreg tapes with final lateral dimensions of 100 mm x100 mm is required for the layup of each test panel. The uncured layup has to be weighed. It is important to mention that the dimensions of the expendable materials have to be sufficiently larger, what results in lateral dimensions of approx. 200 mm x 200 mm. A PTFE-covered perforated foil covers the laminate on both sides. The experimental investigation according to DIN EN ISO 2332 [10] requires three additional layers of glass fiber fabric with a weight of 181 g/m^2 on both sides of the layup, as illustrated in Figure 4, in order to absorb the resin flown out of the prepreg layup during the curing process. A peel-ply is used to remove the laminate after the curing process. In contrast to the requirements according DIN EN 2332 [10], the layup has to be modified by adding a bleeder on the top side in order to make the vacuum bag suitable for cuing in autoclave processing instead of in a heatable press. The test panels are cured in the autoclave with the parameters over the process time as required by the resin's datasheet indicated in Table 3. After curing the layup is unpacked from the expendable materials and a rectangular plate with lateral dimensions of 70 mm x 70 mm is cut out of the center of the test panel. The resin flow in mass-%follows by [10].

$$F_1 = \left(1 - \frac{m_q \cdot s_1}{m_0 \cdot s_2}\right) \cdot 100 \tag{1}$$

where m_0 is the mass of the prepreg layup before curing, m_q is the mass of the cut out rectangular plate, s_1 is the area of the prepreg layup before curing and s_2 is the area of the cut out rectangular plate.

5.4.2. Mass per unit area

The indication of the mass per unit area is required according to the German standard DIN 65090 [21]. Its experimental determination is carried out according to the German standard DIN EN 2329. The mass per unit area in g/m^2 follows by [22]:

$$m = \left(\frac{m_0}{4 \cdot \mathrm{m}^2}\right) \cdot 100 \tag{2}$$

where m_0 is the mass of the prepreg layup before curing, and the factor 4 in the denominator is based on the number of single layers in the layup.

5.4.3. Fiber volume content

The fiber volume content is a significant parameter for mechanically characterizing fiber reinforced plastics. It can only be determined experimentally. In the case of glass and basalt fibers, as anorganic reinforcement fibers, the fiber volume content is determined according to German standard DIN EN ISO 1172 [9]. The polymeric matrix gets evaporated thermally, i. e. is reduced to ashes, whereas the anorganic reinforcement fibers remain due to their high thermal resistance [23]. In detail the specimens are exposed to a temperature of 620 °C for 20 min. By using the density out of the technical data sheet, indicated in Table 3 for the respective reinforcement fibers and in Table 1 for the respective polymeric matrix systems, and the masses of the specimens before and after the calcination process weighted on a precision balance, the fiber volume content φ_f in % is calculated by [9].

$$\varphi_f = \frac{V_f}{V_c} = \frac{V_f}{V_f + V_m} = \frac{\dfrac{m_f}{\rho_f}}{\dfrac{m_f}{\rho_f} + \dfrac{m_c - m_f}{\rho_m}} \tag{3}$$

where the subscripts f, m and c indicate the fiber, matrix an composite properties, respectively, V is the volume, m is the mass and ρ is the density [9, 23, 25]. In order to achieve statistically reliable values eight specimens have been cut out of each test panel and investigated experimentally

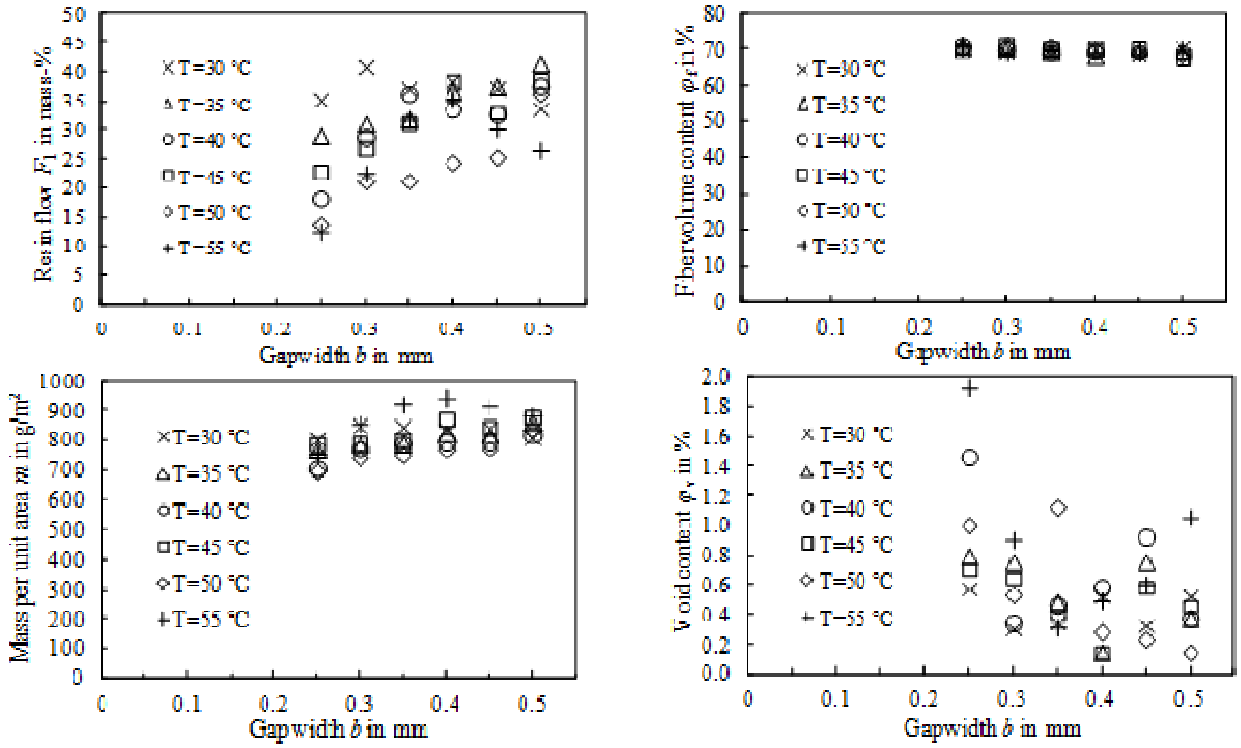

Fig. 4. Prepregs and composite of the material combination E-glass [13,14] and Epikote [16]

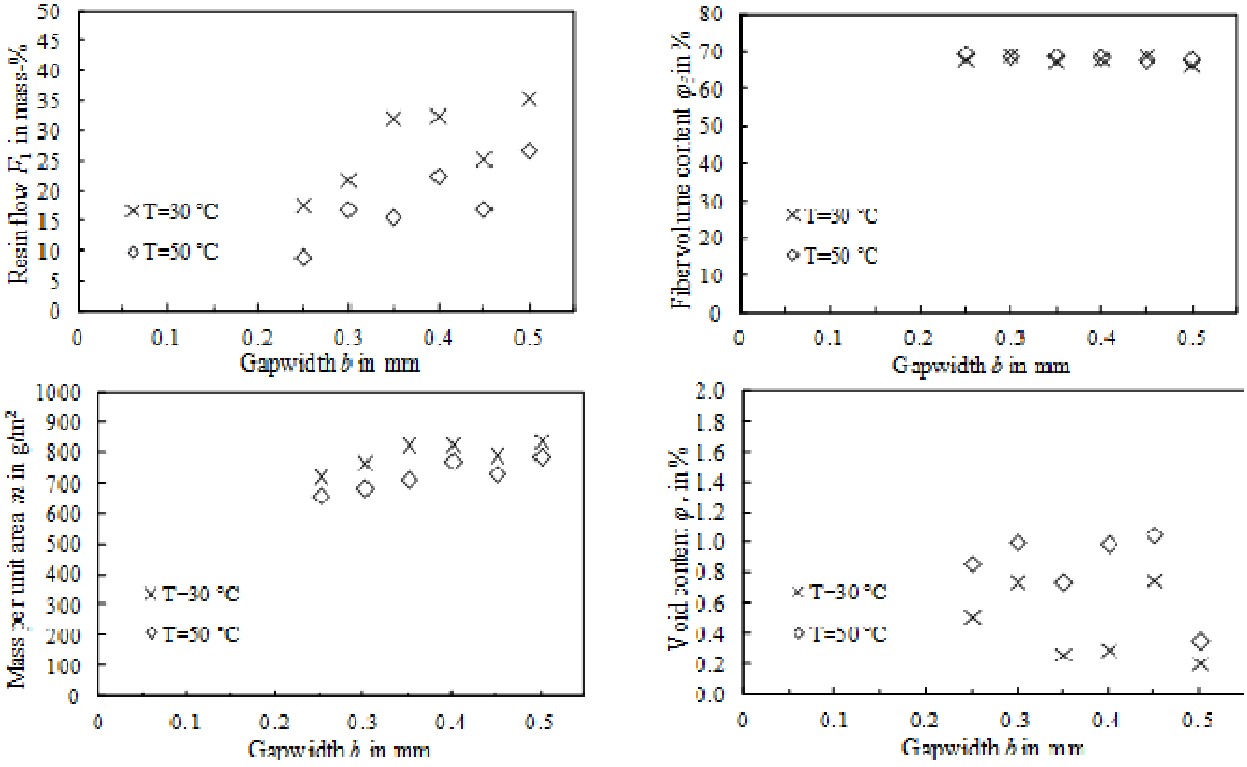

Fig. 5. Prepregs and composite of the material combination E-glass [13, 14] and CeTePox [17]

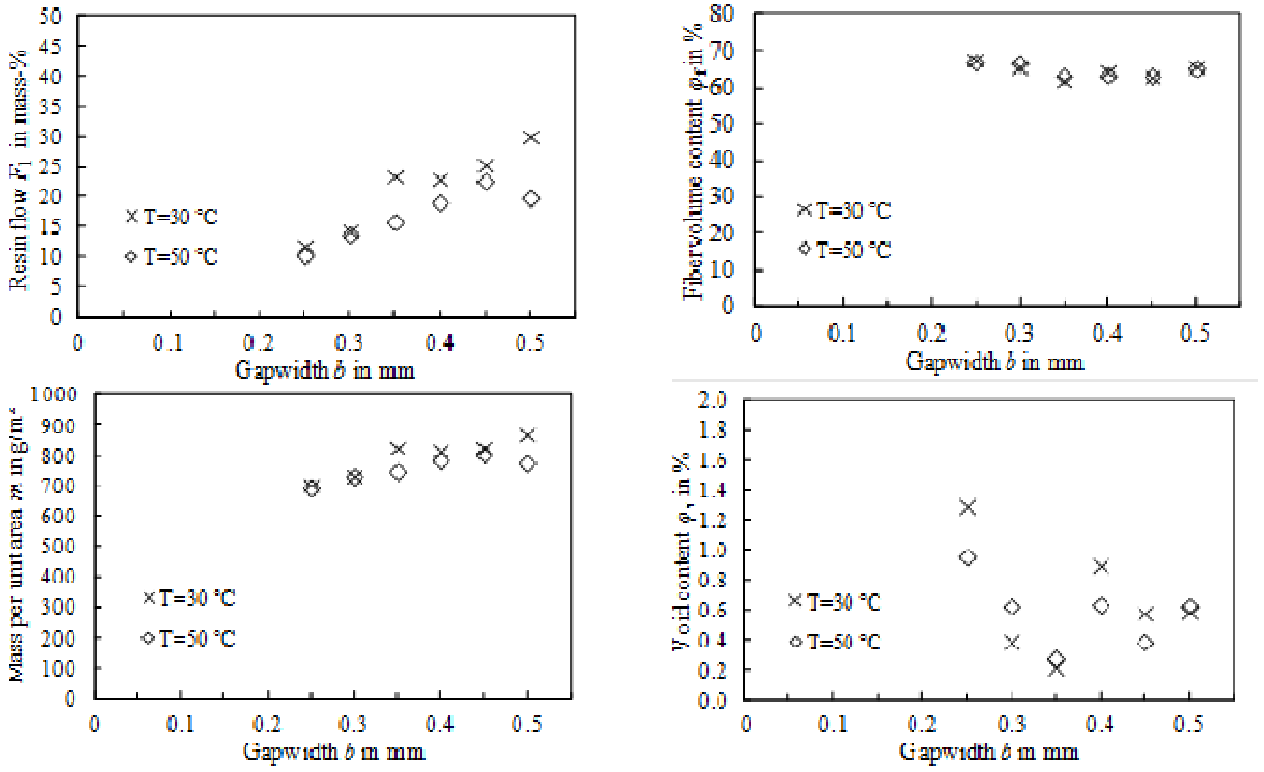

Fig. 6. Prepregs and composite of the material combination E-glass [13, 14] and Sicomin [18]

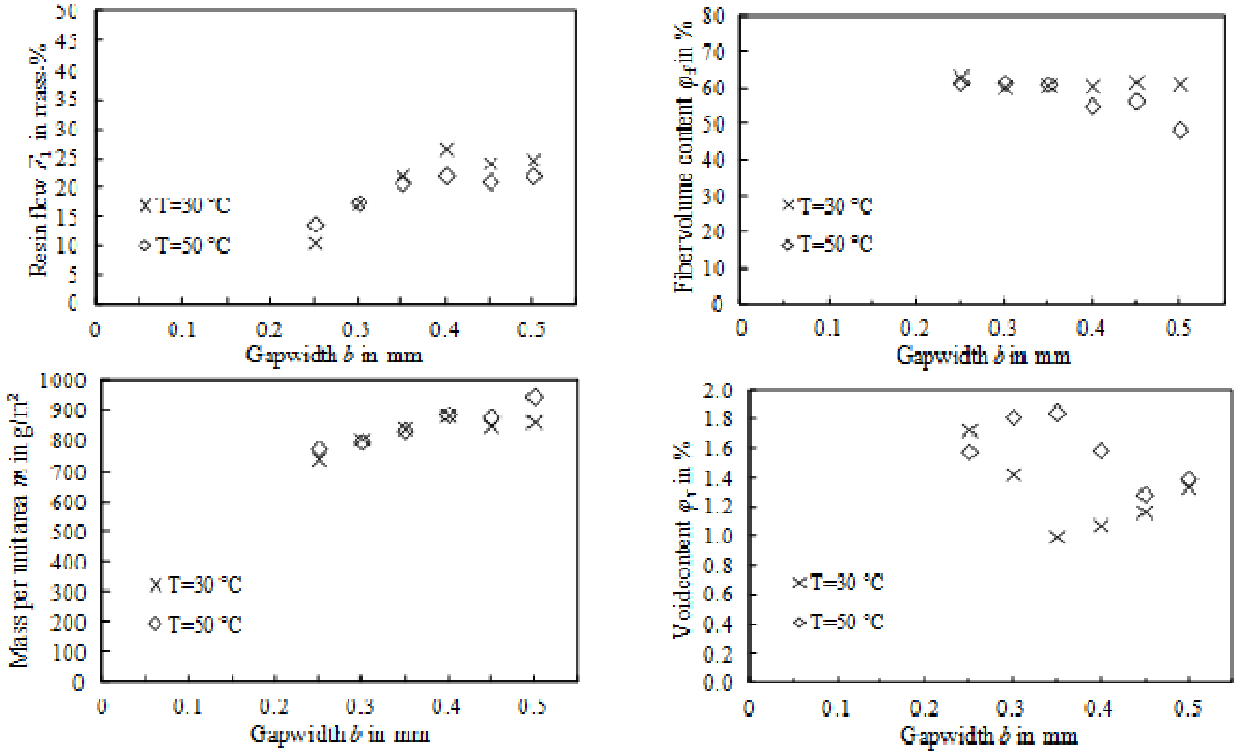

Fig. 7. Prepregs and composite of the material combination basalt [15] and Sicomin [18]

5.4.4. Material density and void content

The material density and the void content are experimentally determined according to the German standard DIN EN ISO 1183 [26]. The physical principle of the experimental determination process is the difference between the lifting forces in two different media. Therefore the mass of the specimens is weighted in air and subsequently in a fluid, here distilled water, whereat the density of both media has to be known. For the determination of the mass of the specimen in the fluid the precision balance mentioned in the previous section 5.4.3. has to be equipped additionally. The density of the specimen, and therefore of the composite material, in g/cm^3 follows by [26]:

$$\rho_{mc} = \left(\frac{m_{S,A}}{m_{S,A} - m_{S,IL}} \right) \cdot \rho_{IL} \tag{4}$$

where $m_{S,A}$ is the mass of the specimen in air, $m_{S,IL}$ is the mass of the specimen in the fluid and ρ_{IL} is the density of the fluid.

The void content in % in the composite material φ_v is evaluated according to the German standard DIN EN ISO 7822 [24]. Thereby the experimentally determined density of the composite material ρ_{mc} according to equation (4) is compared to the theoretical density of the composite material ρ_c.

The theoretical density of the composite material ρ_c results by calculating the weighted average of the densities of the single components, ρ_f indicated in Table 3 for the respective reinforcement fibers and ρ_m indicated in Table 1 for the respective polymeric matrix systems, based on the fiber volume content φ_f, as evaluated in equation (3). The void content in % in the composite material follows by [17].

$$\varphi_v = \left(\frac{\rho_c - \rho_{mc}}{\rho_c} \right) \cdot 100 \tag{5}$$

where ρ is the density, and the subscripts c and mc indicate the theoretical and the experimentally determined value, respectively.

6. Results and discussion

In the following the evaluated results are graphically illustrated and discussed further.

6.1. Results

The evaluated results of the experimental characterization of the prepregs and of the composite material are graphically illustrated. Therefore the evaluated results are plotted against the gapwidth *b*. The results of the four material combinations indicated in Table 4 are illustrated separately in the Figures 4, 5, 6 and 7 and each selected impregnation temperature is considered by a corresponding data series.

In each case the resin flow F_1 in mass-% according to DIN 2332 [10] and evaluated by equation (1) is shown on the upper left, the fiber volume content φ_f in % according to DIN EN ISO 1172 [9] and evaluated by equation (3) is shown on the upper right, the mass per unit area *m* in g/m^2 according to DIN EN 2329 [22] and evaluated by equation (2) is shown on the lower left, and the void content in the composite material φ_v in % according to DIN EN ISO 7822 [24] and evaluated by equation (5) is shown on the lower right.

6.2. Discussion

The following discussion of the obtained results mainly addresses tendencies that can be explained generally.

6.2.1. Resin flow

The consideration of the resin flow plotted over the gapwidth shows the tendency of an increasing resin flow with increasing gapwidth. As the quantity of reinforcement fibers remains constant, the increasing gapwidth provides an increasing quantity of resin remaining in the prepregs. The increasing mass per unit area of the uncured prepregs with increasing gapwidth confirms the described effect. Referring the increasing mass per unit area of the uncured prepregs to the fiber volume content of the cured composite material, there is a nearly constant fiber volume content over the gapwidth. Thus, the additional mass of resin imported with increasing gapwidth has to flow out during the autoclave cycle, in the transformation process from the uncured layup of prepregs to the resulting composite material, in order to achieve a nearly constant fiber volume content over the gapwidth.

The resin flow increases with increasing gapwidth, too, when higher impregnation temperatures are considered. However, the overall resin flow is smaller. This effect can be lead back to the lower viscosity of the resin at higher temperatures, that provides a better impregnation of the single fibers in the rovings during the impregnation

process. Due to the better adhesion between the resin and the reinforcement fibers at higher impregnation temperatures, the tendency to flow out during the autoclave cycle is lower, and more resin remains in the layup. Additionally, at increased impregnation temperatures there is the possibility of a pre-curing effect in the resin, that increases the adhesion between the resin and the fibers, and reduces the resin flow.

The comparison of the E-glass fiber reinforced materials shows, that the resin Sicomin [18] yields the lowest resin flow. As the resin Sicomin [18] provides the lowest viscosity of the three selected polymeric matrix systems as indicated in Table 2, this leads to the conclusion, that a lower viscosity provides a good impregnation of the reinforcement fibers.

6.2.2. Fiber volume content

Considering the fiber volume content of the cured composite materials, especially the E-glass fiber reinforced materials show a nearly constant fiber volume content over the increasing. At first an increasing gapwidth maybe considered providing lower fiber volume contents. The reason therefore is, that during the impregnation process more resin remains in the prepregs, what can be confirmed by the tendencies of increasing mass per unit area of the uncured prepregs over increasing gapwidth. However, the increasing tendency of resin flow with increasing gapwidth shows, that the exceeding resin flows out during the autoclave cycle, in the transformation process from the uncured layup of prepregs to the resulting composite material.

The relatively high values of the fiber volume content partially reach approx. 70 %, because in this state the maximum packing density of the fiber array is reached, and a further compaction of the layup is not possible. In the space between the partially each self contacting fibers the remaining resin resides. The effect of each self contacting fibers yields to a loss in stiffness and strength. In detail at high autoclave pressures the pressure load is increasingly carried by the fibers, and the hydrostatic pressure of the resin decreases. For technical reasonable and applicable values of the fiber volume content values of approx. 60 % have to be achieved. The relatively high values arise from the autoclave pressure of 4 bar indicated in Table 3, due to the simulated heatable press required by DIN EN ISO 2332 [10]. For a possible application of the prepregs for structural parts and load-bearing structures, respectively, the autoclave pressure as a relevant parameter has to be adopted in order to achieve values of the fiber volume content of approx. 60 % or slightly smaller in the composite material after the curing process.

Considering the material combination of basalt fibers [8] and the resin Sicomin [18], a relatively big gapwidth and an impregnation temperature of 50 °C yields relatively low values of the fiber volume content of approx. 55 %. Thereby the resin flow remains relatively low. A possible reason for the relatively good impregnation properties of the basalt fibers and the tendency of the resin in remaining in the laminate is the sizing of the basalt fibers [15]. Besides, the basalt fibers are provided as assembled rovings in contrast to the E-glass fibers, that are provided as so-called direct rovings. Possibly the assembled roving is easier to expand and consequently easier to impregnate in the prepreg production device.

6.2.3. Mass per unit area

As expected, the mass per unit area of the uncured prepregs increases with increasing gapwidth. The impregnation temperature only partially influences the mass per unit area. The material combination of E-glass-fiber [13, 14] and the resin CeTePox [17] in each case exhibits smaller values of the mass per unit area at higher impregnation temperatures as at lower impregnation temperatures.

6.2.4. Void content

In general the void content of all test panels as cured composite in every material combination and for every parameter variation lies in technical reasonable and applicable ranges. Most test panels exhibit void contents smaller than 1 %. Thereby the tendency of decreasing void content with increasing gapwidth and corresponding increasing resin flow is noticeable. On the one hand this can be lead back to an eased leak of the voids from the resin during the curing process. On the other hand the longer availability of sufficiently much resin in the layup keeps the hydrostatic pressure of the resin on a sufficiently high level. The vapor pressure of the soluble parts in the resin is not reached, so that the outgassing is inhibited.

7. Conclusions and outlook

Unidirectionally reinforced prepregs are produced in laboratory scale in a prepreg production device. In detail four material combinations based on an E-glass fiber and a basalt fiber with three kinds of thermoset polymeric matrix systems are investigated. The varied parameters are the impregnation temperature and the gapwidth in the

impregnation module. For an extensive parameter identification and analysis of the sensitivity detailed experimental characterizations of the uncured prepregs and of the cured composite material are carried out. These are for each material combination and each selected parameter variation the resin flow F1 in mass % according to DIN 2332 [10], the fiber volume content φ_f in % according to DIN EN ISO 1172 [9], the mass per unit area m in g/m^2 according to DIN EN 2329 [22], and the void content in the composite material φ_v in % according to DIN EN ISO 7822 [24]. There is a significant dependence between the single varied parameters.

High impregnation temperatures reduce the viscosity of the resin and enhance the impregnation quality. This effect provides a good adhesion between the resin and the reinforcement fibers. This reduces the resin flow during the curing process, and causes the tendency for the fiber volume content to slightly increase. An increasing gapwidth causes an increasing mass per unit area. In autoclave curing process this leads to an increasing resin flow, what causes an eased leak of the voids in the resin. Moreover, greater amounts of resin in the layup inhibit the decreasing of the hydrostatic pressure of the resin below the vapor pressure of the soluble parts, that would cause voids in case of outgassing.

The identified tendencies are clearly evident in the evaluated results of the carried out experimental characterizations of the uncured prepregs and of the cured composite material. Thereby, the basis for the production of independently selectable combinations regarding the single components, namely reinforcement fiber and matrix system, as a prepreg in laboratory scale for research and development issues has been achieved. Improvement potential can be identified in the optimization of the autoclave curing cycles and adaption to the respective prepreg material regarding the achievement of technical reasonable and applicable values of the fiber volume content for a possible application of the prepregs for structural parts and load-bearing structures. Additionally a more exact determination of the afore mentioned pre-curing effect as the so-called B-stage and knowledge about the progress of the reaction in stored prepregs would be useful for generally reducing the fiber volume content.

As different material combinations in terms of reinforcement fibers and polymeric matrix system show different affinity to being impregnated, and even different sizings affect this property, it is likely that another combination of the single components fiber and matrix affects the investigated correlations. In order to investigate the sensitivity of the achieved results on different material combinations further similar investigations with the respective material combinations have to be carried out.

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References

[1] M. Flemming, G. Ziegmann, S. Roth, Advanced composite materials: manufacturing processes with thermoset matrix. Springer-Verlag, Berlin/Heidelberg, 1999.

[2] H. Schürmann, Designing with fiber-plastic composites, Springer-Verlag, Berlin/Heidelberg/New York, 2005

[3] DIN 65378-1 - Preparation of test sheets of unidirectional yarns and rovings. Standards Office Aviation (NL) at DIN Institute for Standardization, e. V., Beuth-Verlag, Berlin/Köln, 1992

[4] R. M. Jones, Mechanics of Composite Materials. 2nd Edition, Taylor and Francis Group, New York, 1999.

[5] M. Flemming, G. Ziegmann, S. Roth, Advanced composite materials: semi-finished products and construction methods, Springer-Verlag, Berlin/ Heidelberg, 1996.

[6] B. Strong, Fundamentals of Composites Manufacturing: Materials, Methods, and Application. Society of Manufacturing Engineers, Dearborn/ Michigan, 1989.

[7] DIN EN ISO 9163 - Textile glass - Rovings - Manufacture of test specimens and determination of tensile strength of impregnated rovings. Normenauschuss Plastics (FNK) in DIN, Textiles and Textile Machinery Standards Committee (textile standard) in DIN, Standards Committee Aerospace (NL) at DIN German

Institute for Standardization e. V., Beuth-Verlag, Berlin, 2005.
[8] A. Giessmann, Substrate and coating textiles: practical knowledge for coating and laminating technologies, Springer-Verlag, Berlin/Heidelberg, 2010.
[9] DIN EN ISO 1172 - Textile glass reinforced plastics - Prepregs, molding compounds and laminates - Determination of the textile-glass and Mineral-füllstoffgehalts - calcination. Plastics Standards Committee (FNK) at DIN Institute for Standardization e.V., Standards Office of Aviation (NL), Beuth-Verlag, Berlin, 1988.
[10] DIN EN 2332 - Glass filament prepreg - Test method for the determination of urinary flow. Standards Office Aviation (NL) at DIN Institute for Standardization, e. V., Beuth-Verlag, Berlin, 1993
[11] DIN 65060 - Textile glass filament rovings for reinforcement of plastics - Technical delivery conditions. Standards Office Aviation (NL) at DIN Institute for Standardization, e. V., Beuth-Verlag, Berlin/Köln, 1991.
[12] E-glass direct roving 076 PR 2400-01. Technical data sheet, Mühlmeier GmbH & Co. KG, Bärnau, 2014.
[13] Glassfiber roving 076 PR 2400-01. Quality certificate, Johns Manville, Slovakia, 2008.
[14] Basalt rovings BR130.2400.12RAA. Assembled standard rovings. Technical data sheet, Incotelogy Ltd., Pullheim, 2013.
[15] Epikote Resin 05128, Epikure Curing Agent 3280 - Hot curing epoxy resin. Technical data sheet, Momentive, 2010.
[16] CeTePox AM VP 382-1 A/B1/B2. Technical data sheet, CTP Advanced Materials GmbH, Rüsselsheim, 2005.
[17] Si Preg 8500. 2-component prepreg-epoxy-system. Technical data sheet, Time Out Composite oHG, Bornheim-Sechtem, 2009.
[18] A.R. Bunsell, J. Renard: Fundamentals of fibre reinforced composite materials. MPG Books Ltd, Bodmin/Cornwall, 2005.
[19] G.W. Ehrenstein, Composites: materials - processing – properties, Carl Hanser Verlag, München/Wien, 2006.
[20] DIN 65090 - Textile glass - Preimpregnated filament of E-glass (prepreg) - Technical specification. Standards Office Aviation (NL) at DIN Institute for Standardization, e. V., Beuth-Verlag, Berlin/Köln, 1.979.
[21] DIN EN 2329- Aerospace - glass filament prepreg - Test method for determining the mass per unit area. Standards Office Aviation (NL) at DIN Institute for Standardization e. V., Beuth-Verlag, Berlin, 1993.
[22] V. Schmid, B. Jungbauer, M. Romano, I. Ehrlich, N. Gebbeken, Diminution of mass of different types of fibre reinforcements due to thermal load, Proceedings of the Applied Research Conference, Nürnberg, 2012, 231-235.
[23] DIN EN ISO 7822 - Textile glass reinforced plastics - Determination of the amount of existing voids - Loss on ignition, mechanical disintegration and statistical analysis methods. Plastics Standards Committee (FNK) at DIN Institute for Standardization e.V., Standards Office of Aviation (NL) im DIN, Beuth-Verlag, Berlin, 2000.
[24] C. Cherif, Textile materials for lightweight construction: techniques - methods - materials - properties, Springer-Verlag, Berlin/Heidelberg, 2011.
[25] V. Schmid, B. Jungbauer, M. Romano, I. Ehrlich, N. Gebbeken, The influence of different types of fabrics on the fibre volume content and porosity in basalt fibre reinforced plastics, Proceedings of the Applied Research Conference, Nürnberg, 2012, 162-165.
[26] DIN EN ISO 1183-1 - Plastics - Methods for determining the density of non-cellular plastics - Part 1: Immersion method, liquid pyknometer method and titration method. Plastics Standards Committee (FNK) at DIN Institute for Standardization, e.V., Standards Office of Aviation (NL), Beuth-Verlag, Berlin, 2013.