GETTING BETTER PROCESS PARAMETERS WITH THE MULTISCALE MODELLING OF THE STABILIZATION PROCESS OF PAN-PRECURSORS DURING CARBON FIBER MANUFACTURING

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Abstract
Carbon fibres combine excellent mechanicals properties with low density and are therefore very often used in lightweight constructions. The production of carbon fibres is divided into three parts. First part is the production of the raw material (precursor). The second is the thermal conversion to stabilize the fibre and prepare it for the final step. In the last part the carbonization almost all but carbon atoms are eliminated. Resulting in a fibre material containing over 90% of carbon. Even though carbon fibre has been used since the late 1970s in aerospace application, there is still no general method available to find the optimal production parameters and the trial-and-error approach is most often the only resolution. Therefore, the stabilization process is analysed experimentally and by simulation. With the presented model it is possible to perform a complete simulation of the fibre undergoing all zones of stabilization. The fiber bundle is modeled as several circular fibers with a layer of air in-between. Two thermal mechanisms are considered to be the most important: the exothermic reactions inside the fiber and the convective heat transfer between the fiber and the air. The exothermic reactions inside the fibers are modeled as a heat source. Differential scanning calorimetry measurements have been performed to estimate the amount of heat of the reactions. To shorten the required time of a simulation the number of fibers is decreased by similitude theory. Experiments were conducted to validate the simulation results of the fibre temperature during stabilization. The experiments for the validation were conducted on a pilot scale stabilization oven. To measure the fibre bundle temperature a new measuring method is developed. The comparison of the results show that the developed simulation model gives good approximations for the temperature profile of the fibre bundle during the stabilization process.

1. Introduction

Because of their excellent mechanical properties, combined with low density, carbon fibres are mainly used in lightweight constructions [1]. For now, the usage of carbon fibres is limited by the complicated manufacturing process and the related high price. The production of carbon fibres consists of three steps. In the first step the raw material (precursor) is produced. Afterwards the produced precursor bundle is treated thermally by temperatures about 250°C to stabilize it for the final step. Therefore, this production step is called stabilization. During the last part, the carbonization, the fibre bundle is treated thermally once again. Heating up the fibre bundle to temperatures up to 1500°C results in a fibre containing over 90 % of carbon atoms. Especially the stabilization process requires a long

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processing time and is thus mainly responsible for the high price of the final carbon fibre. To get the best performing during stabilization each precursor requires a suitable temperature profile. Currently these profiles are determined by the method of trial and errors. Therefore the stabilization process is scaled down to laborious test runs. Using a simulation model is a promising method to predict the resulting stabilization of a precursor without spending much time and money in laborious test runs [2].

2. Methodology

The presented model is used to simulate the way of a fibre bundle through a stabilization oven by a two dimensional transient thermal analysis. The fibre bundle is modelled as several circular fibres with a layer of air in-between (Fig. 1).

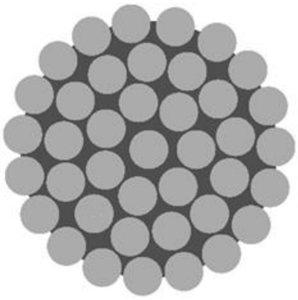

Figure 1. Assumed arrangement of circular fibres

Two thermal mechanisms are considered to be the most important: the exothermic reactions inside the fibre and the convective heat transfer between the fibre and the air. The convective heat flow is calculated by equation (1) using a Nusselt-Correlation [3].

$$\dot{q}'' = \alpha \cdot (T_{Fiber} - T_{Air}) \tag{1}$$

$$\alpha = \frac{0{,}84 \cdot \lambda_F \cdot Re^{0{,}334}}{d} \tag{2}$$

The calculation of the reaction heat flow is based on a new approach using DSC-Measurements. The reaction heat flow is calculated by (Eq. 3) and consists of two factors: ${}^{c}/_{c_0}$ and $\dot{H}_{Reaction,0}$

$$\dot{H}_{Reaction} = \frac{c}{c_0} \cdot \dot{H}_{Reaction,0} \tag{3}$$

The first factor describes the decreasing concentration of the reactant and therefor the thermal degradation of the PAN-Precursor. Due to the lack of data about the concentration of the reactant, this factor is calculated by equation (4) using the released reaction enthalpy and releasable reaction enthalpy. The releasable reaction enthalpy comprises the amount of energy that can theoretically be released during the stabilization process and varies with the precursor. DSC-Measurement have been performed to determine the value.

$$\frac{c}{c_0} = 1 - \frac{H_{released}/_{m}}{H_{releasable}/_{m}} \tag{4}$$

The second factor describes the temperature dependence of the exothermal reactions by a DSC-Measurement. The measured heat flow is a function of the temperature and the concentration of the reactants, latter is already included in the first factor. Thus, the values needs to be adjusted by equation (5), due to the decreasing concentration of the reactants during the measurement (**Fehler!**

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$$\dot{H}_{Reaction,0} = \frac{\dot{H}_{DSC}}{c/c_0} \tag{5}$$

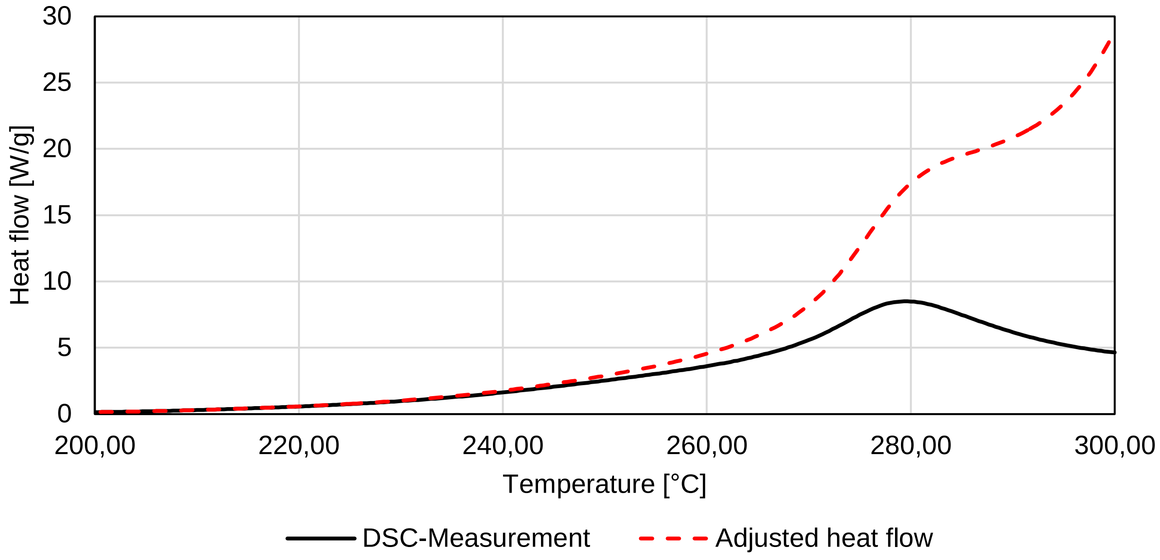

Figure 2. Adjusted heat flow

The simulation of a fibre bundle with a few thousand fibres is time consuming. To shorten the required time of a simulation, the number of fibres is decreased by similitude theory. Three dimensionless quantities are used:

$$\theta = \frac{convective\ heat\ flow}{reaction\ heat\ flow} \tag{6}$$

$$Bi = \frac{\alpha \cdot d_{Bundle}}{\lambda_S} = \alpha \cdot d_{Bundle} \cdot \left(\frac{P}{\lambda_{PAN}} + \frac{(1-P)}{\lambda_{Air}}\right) \tag{7}$$

$$P = \frac{n \cdot r_{Fibre}^2}{r_{Bundle}^2} \tag{8}$$

The first dimensionless quantity leads to equation (9), which is used to calculate the heat transfer coefficient of the model. As the equation shows, α_{Model} depends on the original heat transfer coefficient and the number of fibres of the original bundle and its model.

$$\alpha_{Model} = \alpha_{Original} \sqrt{\frac{n_{Model}}{n_{Original}}} \tag{9}$$

By equation (7) the thermal conductivity of the model is modified. As a result of this the temperature distribution within the bundle of the original bundle and its model is similar. The geometric similarity is obtained by the packing density P, which describs the ratio of fibre material to air.

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3. Results

The data used for the model validation is obtained from an experiment. The temperature of the fibre bundle is measured during the stabilisation process. The process parameter are detailed in **Fehler! Verweisquelle konnte nicht gefunden werden.**. The results of the experiment and the simulation are shown in **Fehler! Verweisquelle konnte nicht gefunden werden.**

Table 1. Process parameter of the experiment and the simulation

Parameter	Zone 1	Zone 2	Zone 3	Zone 4
Temperature [°C]	200	220	240	260
Dwell time [min]	19,4	19,4	19,4	19,4

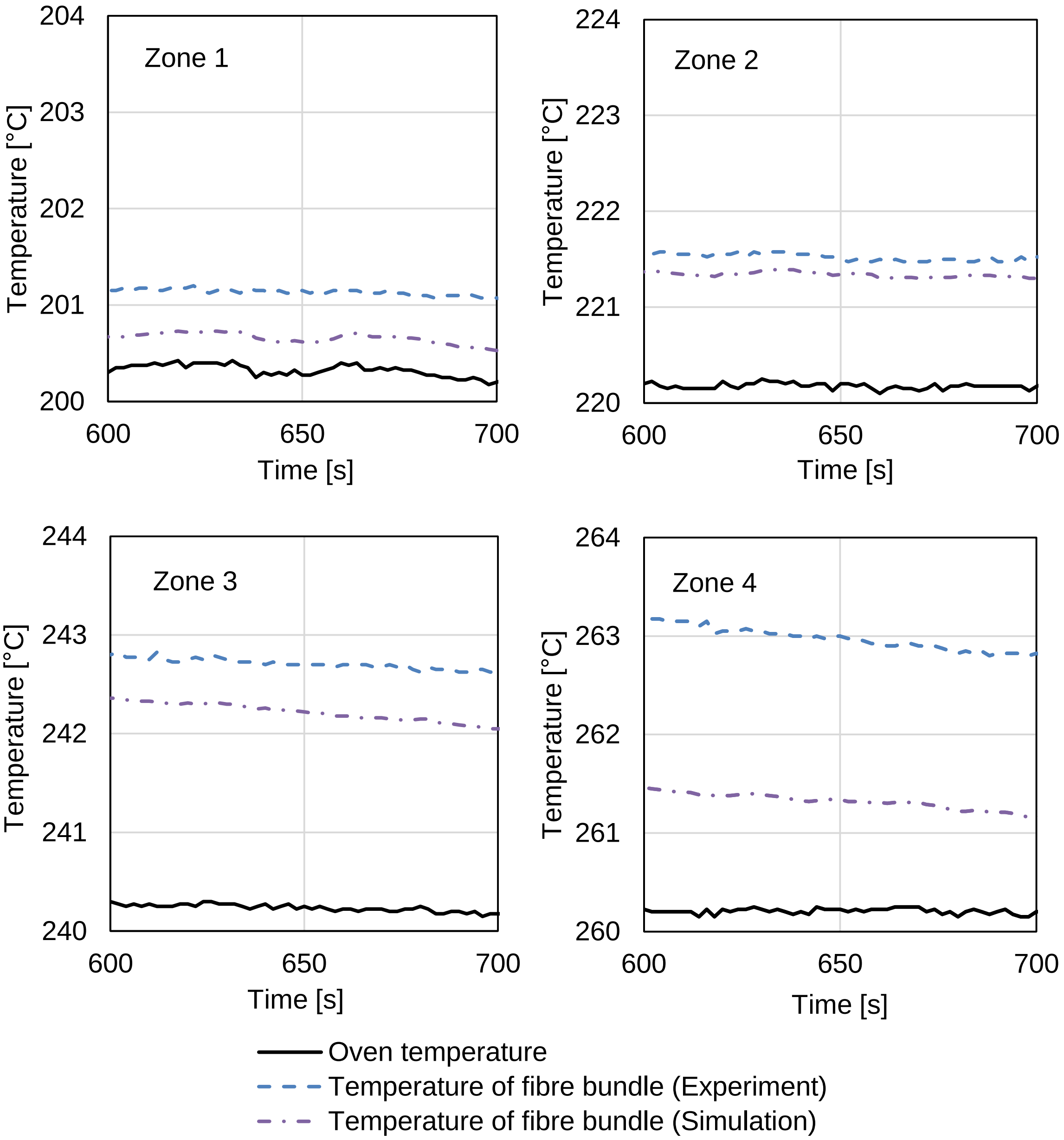

Figure 3. Measured and simulated temperature of the fibre bundle

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The results show, that the temperature of the simulation is above the oven temperature but below the temperature of the experiment in every temperature zone. The temperature difference between the results of the experiment and the simulation varies with the temperature zone. In the second and third zone the difference is lower than the difference in the first and fourth zone. The temperature difference in the second and third zone is negligible. The new approach to calculate the reaction heat flow shows good results. A drop of the temperature, due to decreasing reactant concentration, can be seen in zone 3 and zone 4.

4. Conclusions and recommendations

The presented simulation is capable to predict the temperature of the fibre bundle during the stabilization process. The presented reaction model appears promising to calculate the reaction heat flow, but additional studies are needed. The heat flow at low temperatures or long processing time seems too low. However, the temperature difference between the experiment and the simulation is not significant. Thus, the presented model can be used to prevent overheating of the fibre bundle and to reduce the energy and time consumption of the manufacturing process.

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