Combination of Scattering Matrix Code and Process Model to Optimize a Microwave Applicator Suitable for the Stabilization of PAN Fibers

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***Abstract*— Carbon fiber production is an energy intensive process requiring new approaches for energy efficient heating. One possible option might be the dielectric heating. A basic requirement to design an efficient applicator is the knowledge of the variation of dielectric properties during processing. The experience shows strongly increasing dielectric loss of a Polyacrylonitrile (PAN) fiber with increasing temperatures while it decreases during the chemical transformation in the stabilization stage. For the applicator design an electrical field that counteracts the variation of the dielectric loss is a suitable choice. In this presentation the focus is on the combination of the generalized scattering matrix (GSM) code with a process model. It shall allow for the optimization of the geometry of a cylindrical resonator usable during the stabilization stage of the PAN fiber. The scattering matrix code is utilized to calculate the field profile of a cylindrical resonator with step-wise changing diameter that acts as applicator. The number of steps can be varied, depending on the ability of production and spacial requirements.**

***Keywords*— Matrix Scattering Code, Resonator, Modelling, Dielectric heating.**

I. Introduction

Lightweight applications benefit from the use of carbon fibers due to their good tensile strength to density ratio. But the production costs are still rather high and the final energy efficiency in the production process of the raw material, the carbon fiber, is far from optimum today. The production stage starts from the polymerization of the material, goes on to the spinning and continues with the stabilization of the fiber at temperatures between 200 °C and 300 °C and the carbonization at temperatures higher than 1000 °C, before finalizing with the post processing, to prepare the fiber for carbon fiber composites. Different precursor materials are used for the production but polyacrylonitrile (PAN) fibers is the most commonly used precursor material [1][2]. Of course each processing step can be optimized to reduce the production costs, but the focus of this work is on the stabilization stage. The fiber is stabilized in air by passing convection ovens at temperature stages ranging from 200 °C up to 300 °C. The process time varies up to several hours depending on the temperature steps, the diameter of the precursor and its chemical composition. The fiber is chemically changed through oxidation, cyclization and dehydration. All three reactions are exothermic. It requires to control the process carefully to avoid an overheating of the fiber. In order to shorten the time and increase the process efficiency, different approaches are investigated such as optimizing the heat distribution and insulation of conventional ovens [1][2]. Improving the stabilization is also investigated by setting up models of the chemical reaction and the development of the temperature [3]. Also replacing the conventional heating by plasma or microwave heating in at least one of the process steps is explored. Microwave heating offers volumetric heating, selective heating and fast temperature control. Microwave heating was so far not included in the modelling of the stabilization. Adding the microwave heating to calculate the temperature development can help in controlling the process and avoid the thermal runaway reaction. The patents EP2475812B1 and US20120137446A1 by Toho Tenax Europe GmbH describe a microwave applicator which uses a combination of a preheated process gas in combination with microwave heating to stabilize the fiber. The microwave applicator consists of areas with different electric field strengths. It consists of a cylindrical waveguide and has a physical length of 1.5 m.

Any appropriate microwave applicator used for dielectric heating is dependent on the behavior of the dielectric loss of the material, which is, obviously, dependent on the chemical composition. In order to design such a suitable applicator, the process model which calculates the temperature of the fiber is combined with a scattering matrix code to calculate the electrical field profile in resonators depending on the dielectric loss. First the adaption and optimization of the scattering matrix code is investigated individually.

II. Generalized Scattering Matrix Code

This section shortly explains the implementation of the scattering matrix code assuming circular waveguides [4]. Any waveguide structure can be approximated as a series of straight waveguide segments that are intersected by step discontinuities. As the transversal electric (TE) and transversal magnetic (TM) eigenmodes of a waveguide form a complete set, any field configuration inside a straight waveguide segment can be described as a superposition of that eigenmodes. In the generalized scattering matrix approach each of these segments is described by its own scattering matrix. In practice only a few eigenmodes are above cut-off at the operating frequency and all other waveguide modes are far below cut-off. Those higher-order eigenmodes are strongly attenuated and can be neglected when combining two scattering matrices. Hence, the scattering matrices of the straight waveguide sections only need to describe the propagation of the modes according to the segment length and each mode's propagation constant. The scattering matrices of the discontinuities on the other hand need to include the coupling between the different modes. For some special cases such as the step between two axially aligned circular waveguides the scattering matrix of the discontinuity can be described analytically which creates the possibility for very fast simulation. An exemplary step discontinuity of a circular waveguide structure in a 2D cut can be seen in Fig. 1, where R_{I} and R_{II} are the radii of the respective waveguide segments, F_{I} and F_{II} the forward traveling waves in the segments and B_{I} and B_{II} the backwards traveling waves.

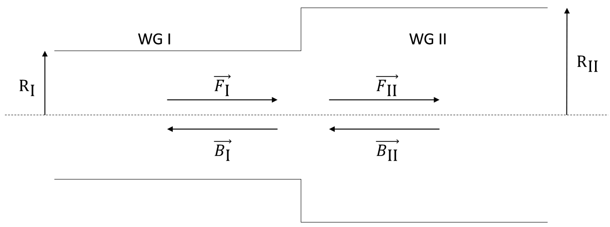


Fig. 1. Step discontinuity of a circular waveguide

In this case the mode matching technique was used to calculate the coupling between the modes at the discontinuity. It relies on the modal decomposition of the electric and magnetic fields inside both waveguides. Therefore the derivation of the eigenmodes inside a circular waveguide is the first step in the development of the simulation approach. Based on solutions for the eigenmodes the formulations of the generalized scattering matrix method and the mode matching method can be formed. The electric and magnetic fields in a circular waveguide can be written as [5]:

$$\vec{E}_I = \sum_{i=1}^{\infty} [F_i + B_i]\, \vec{E}_{I,i} \tag{1a}$$

$$\vec{H}_I = \sum_{i=1}^{\infty} \left[\frac{F_i - B_i}{Z_i}\right] \vec{H}_{I,i}, \tag{1b}$$

where F_i and B_i are the elements of a vector containing the amplitude and phase of the forward and backward traveling waves, Z_i is the wave impedance and $\vec{E}_{I,i}$ and $\vec{H}_{I,i}$ are the electric and magnetic fields of the i-th mode inside the waveguide I. Equally the field in the second waveguide can be defined. At the mutual cross section of both waveguides, the tangential E and H fields have to satisfy the continuity law, which leads to:

$$[P][F_{\mathrm{I}} + B_{\mathrm{I}}] = [I][F_{\mathrm{II}} + B_{\mathrm{II}}] \tag{2a}$$

$$[Z_{\mathrm{I}}][P]^t[Y_{\mathrm{II}}][F_{\mathrm{II}} - B_{\mathrm{II}}] = [I][F_{\mathrm{I}} - B_{\mathrm{I}}] \tag{2b}$$

where $[Z_{\mathrm{I}}]$ is a diagonal matrix containing the wave impedances in the first waveguide and $[Y_{\mathrm{II}}]$ is a diagonal matrix containing the wave admittances in the second waveguide.

The matrix $[P]$ contains the mode coupling coefficients which are defined by the integral

$$P_{ij} = \int_{s_{\mathrm{I}}} \vec{E}_{j,\perp,\mathrm{II}} \cdot \vec{E}_{i,\perp,\mathrm{I}} \mathrm{d}s_1, \tag{3}$$

with the tangential electric fields $\vec{E}_{j,\perp,\mathrm{I}}$ and $\vec{E}_{j,\perp,\mathrm{II}}$ in both waveguides. The existing scattering matrix code for circular waveguides was extended to allow the calculation of closed resonators following the procedure presented by Neilson et al. [5]. This requires the structure to be split in two parts, which are both terminated in a short. The scattering matrices of both parts are then cascaded into the single scattering matrices S_{11}^R and S_{11}^L. The condition for resonance inside the cavity is that the forward and backward traveling waves at the split point are equal. This leads to the eigenvalue equation:

$$\vec{B^R} = S_{11}^R \cdot S_{11}^L \cdot \vec{B^R} \tag{4}$$

which is solvable if the system is resonant at the current frequency. After the resonance frequency has been found, the corresponding eigenvector $\vec{B^R}$ can be used to calculate the amplitudes in the other resonator segments using the scattering matrices of the individual segments. When the mode amplitudes in all segments are known, the electric field profile can be calculated for each segment using equation (1a).

III. Process Model

In this section the setup of the process model is explained as well as the temperature dependent results of dielectric measurements of a PAN precursor are given. The knowledge of the dielectric loss is the base for calculating the power absorbed inside the material which leads to an increase in the temperature and thus for optimizing the field pattern later on.

A. Dielectric Properties

Mimicking the conventional process as close as possible, a heating rate of 30 °C/min was chosen for all measurements. As heating rate, final temperature and holding time have a great influence on the chemical reaction, they also affect the dielectric properties. In Fig.2 the measurement results can be seen for a process temperature of 260 °C and 50 min holding time. More results and details to the measurement system can be found in [6]. The process phase is divided in the heating up phase (blue), holding at the final temperature (red) and cooling down phase (yellow). A strong temperature dependency of the dielectric loss factor is visible for the heating up phase. It can be said that for a virgin PAN fiber the loss factor rises two orders of magnitude with increasing temperature. It decreases again with the ongoing chemical reaction that unfolds during the holding time.

B. Setup

A thermal model helps to built an understanding of microwave heating processes and can be a first step towards online monitoring and controlling of a continuous process. Awareness of the influence of the process parameters can help predefine the input variable range which leads to the

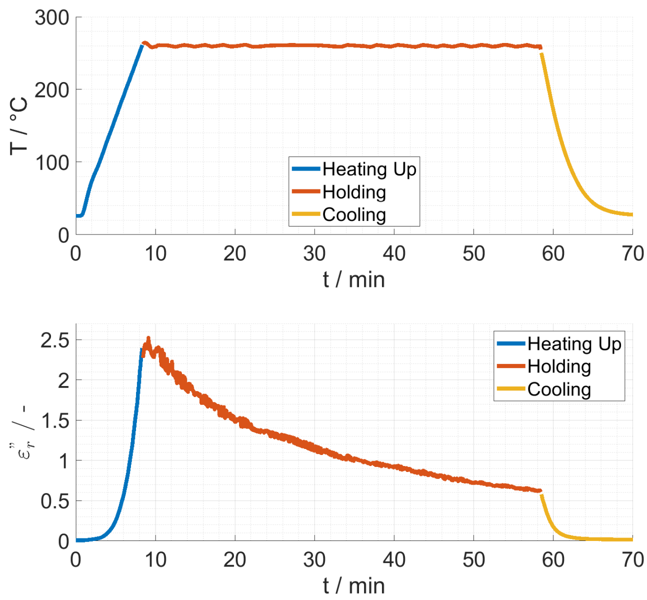


Fig. 2. Change of dielectric loss

desired output temperature instead of a thermal runaway. The model calculates the temperature rise due to the heat induced by the microwaves, the time inside the microwave system, the reaction heat from the exothermic reactions and the heat transfer. The temperature at every z-position along the microwave length can be calculated and thus used to predict the behavior of the fiber during the reaction. As the setup is symmetrical, only a 2D Model was implemented of the inner part: the PAN fiber within the quartz tube. The 2D model comprises multiple mesh cells in axial and radial direction. The number of mesh cells can be varied in both directions. This enables the user to find the optimum between computation time and result accuracy. Each cell is connected to the adjacent cells in axial and radial direction via the thermal equations. The calculation of the thermal equations was implemented in MATLAB by Mathworks® and solved for each cell. The change in temperature T can be calculated by [7]:

$$\frac{dT}{dt} = \frac{\dot{Q}_{sum}}{m \cdot c_p} \tag{5}$$

where Q_{sum} is the sum of all heat sources, m the mass of the material and c_p the specific heat capacity of the material. In Q_{sum} the convection $\dot{Q}_{conv}$, conduction $\dot{Q}_{cond}$,radiation $\dot{Q}_{rad}$, the heat released by the exothermic reactions $\dot{Q}_{reac}$ as well as the heat induced through microwaves is included:

$$\dot{Q}_{sum} = \dot{Q}_{conv} + \dot{Q}_{cond} + \dot{Q}_{rad} + \dot{Q}_{reac} + p_{mw,V} \cdot V \tag{6}$$

where V is the volume of the cell. The heat through microwaves is calculated as follows [7]:

$$p_{mw,V} = 2 \cdot \pi \cdot f \cdot \varepsilon_r'' \cdot \varepsilon_0 \cdot |E|^2 \tag{7}$$

where f is the frequency, ε_r'' the dielectric loss, ε_0 the vacuum permittivity, and E the electrical field strength.

The heat flow obtained through the chemical reactions $\dot{Q}_{reac}$ is based on the reactions kinetics evaluated from the dielectric point of view in combination with the heat of reaction evaluated for two reactions, the cyclization and the oxidation, by the broken and formed bond enthalpies [8].

IV. Discussion

The scattering code can not only be used in combination with the process model, but also individually to optimize geometries for arbitrary electrical fields. The optimizer uses boundaries for the cavity length, the length and the radius of the segments, the frequency and has the option to either predefine a fixed number of segments or use a variable number of segments. This allows to either keep in mind the manufacturing process or to explore the theoretical optimal geometry.

A. *Optimization of an arbitrary electrical field*

Exemplary the following equation (eq. 8) was used to describe an arbitrary field profile along the z-axis at the point R=0 mm:

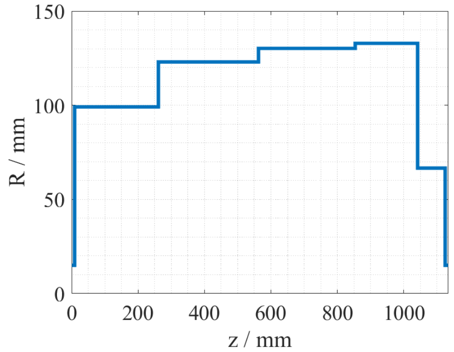
$$f(x) = sin(z) \cdot e^{-z} \tag{8}$$

The optimized geometry cross section can be seen in Fig. 3a with the comparison between the desired function and the optimized result in Fig. 3b. The optimized function is not yet equal to the desired field profile, but this can be improved when more steps are used for the optimization.

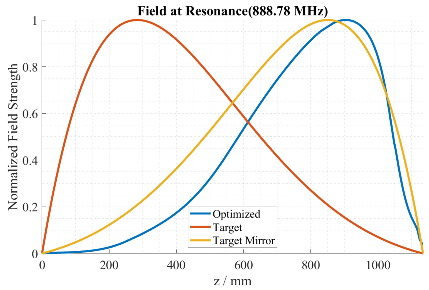
B. *Optimization of desired temperature profile*

In a first step the heating up of the fiber was used as goal temperature profile to find a suitable electrical field based on a fixed number of sampling points independently from the scattering code. The electrical field was setup by optimizing four sampling points and linearly interpolating in between. A comparison of the exemplary goal temperature profile and optimized profile can be seen in Fig. 4, which show a good enough agreement to go on with the optimization of the geometry.

In a second step the GSM code optimizer was given the obtained sampling points as input to optimize the geometry of the applicator. The utilized boundaries are chosen as follows: cavity length fixed at 40 cm, segment length between 1 and 30 cm, segment radius between 2 and 15 cm and frequency between 700 and 1100 MHz. The sampling points, obtained from the Process Model, are used to describe the electrical field profile along the z-axis at the point R=0 mm by interpolating in between. In Fig. 5a the optimized 2D axial symmetrical profile with four segments can be seen with the comparison of the respective electrical field profile in Fig. 5b to the desired electrical field. It can be seen that with four segments a rather good agreement can be achieved.



(a)



(b)

Fig. 3. Optimization of arbitrary field profile with 5 segments : (a) Geometry; (b) Field Profile

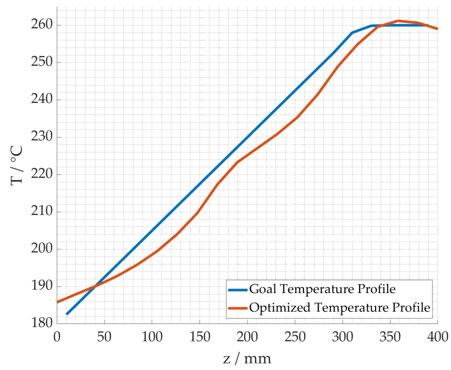
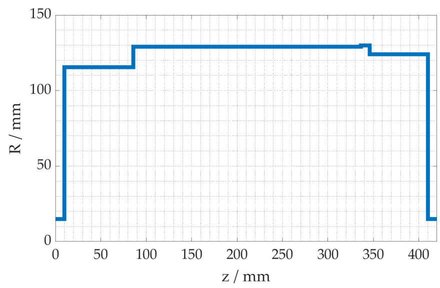


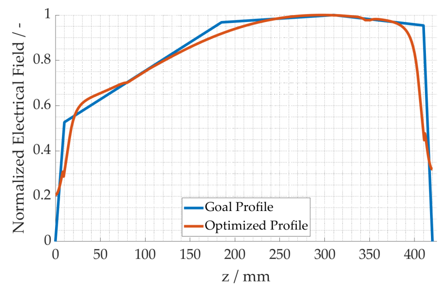
Fig. 4. Comparison goal and optimized temperature profile

V. Conclusion

The efficient utilization of the scattering matrix code together with a model of the reaction kinetics for the optimization of a microwave applicator for effective dielectric heating of carbon fibers during the stabilization phase is discussed. Using the scattering matrix code, the manufacturing possibilities and the spacial requirements have to be kept in mind during the optimization. The next steps will be to use the full desired temperature profile which is needed for the complete stabilization as goal function for the optimization of the resonator geometry. Further possible next steps can be to extended the scattering matrix to be able to also calculate the quality factor of the system.



(a)



(b)

Fig. 5. Optimization of electrical field profile with 4 segments based on sampling points: (a) Geometry; (b) Field Profile

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