Fabrication and upscaling of spinning processes for ceramic high-tech fiber production

Ceramic fibers for high-tech applications require intensive developments concerning precursor synthesis and fiber spinning. The spinning process starts at laboratory scale with only few filaments and needs to be upscaled to pilot plant and industrial scale with hundreds or even thousands of filaments. Due to the significant interaction of filaments and surrounding inert atmosphere, the behavior in the spinning tower completely changes with higher numbers of filaments. How these effects are incorporated in a model of the gas flow including its influence by the filament spinning will be described. In 2 examples for melt- and dry-spinning of ceramic fibers it will be demonstrated how characteristic properties derived from experiments on a laboratory scale can be efficiently transferred to a pilot plant scale by use of simulations. For both applications the spinning tower was built according to the conditions derived from the simulation results, and experimentally proven to work.

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Ceramic fibers can be grouped according to their chemical composition into oxide and non-oxide fibers. Examples are Al_2O_3, $3\,Al_2O_3$-$2\,SiO_2$, SiO_2 for oxide and SiC, Si(B)NC and C for non-oxide ceramic fibers. Ceramic fibers are characterized by high tensile strength and stiffness, high temperature stability and a good corrosion resistance. They are used in applications like reinforcement of ceramic or metallic composites (CMC: ceramic matrix composites; MMC: metal matrix composites), insulation, filtration and catalysis.
Ceramic fibers for reinforcing composites are typically produced by a melt- or dry-spinning process. During the development of ceramic fibers, the upscaling from laboratory to pilot plant scale is associated with high costs due to the need of extensive experimental studies and expensive equipment.
The use of modern simulation tools in combination with experimental work can provide possibilities to develop ceramic fibers more cost-efficiently.

Requirements for the fiber spinning processes

The concept is demonstrated by 2 applications, the melt-spinning and the dry-spinning of Si-polymers, which are used to fabricate non-oxide ceramic fibers. In both cases inert process conditions are realized by flange-mounting glove boxes on the ends of the spinning tower, so that the oxygen absorption of the spun filaments before the final pyrolysis step is as low as possible. During the pyrolysis process at temperatures above 1,000 °C, the polymer filaments are transformed to ceramic filaments by decomposition and evaporation of organic species. For the melt-spinning of Si-polymers, the polymer filaments need to be cured in order to prevent re-melting during the pyrolysis. A first curing step of the spun polymer filaments is already realized in the spinning tower by the controlled metering of curing gas. The supplied curing gas needs to show a homogeneous distribution in the spinning tower except the glove boxes in order to cure all filaments in the bundle completely. In contrast to melt-spinning, for the dry-spinning process Si-polymers with a different degree of cross-linking are used. In this case, a curing step is omitted. The solvent needs to be removed from the spun filaments in a controlled manner in order to reach a minimal defect level. Therefore, solvent in vapor form is supplied to the spinning tower with the challenge of realizing a concentration gradient in vertical direction of the spinning tower to allow a nearly complete removal of the solvent from the spun filaments. In both cases it is important that the supplied gases do not propagate into the glove boxes mounted at the spinning tower. It is essential to exactly control the gas supply to achieve a high and reproducible fiber quality. It is observed that the gas flow and distribution are significantly changed during upscaling to a larger filament number. Finding the optimal process parameters for each stage of upscaling by mere experimental studies is almost impossible.

Modeling and simulations

A crucial point for the upscaling of these spinning processes is the surrounding gas flow. Its modeling is based on the conservation laws for mass, momentum and energy (Navier-Stokes equations). The mass balance includes not only the inert gas but also reactants or solvents depending on the specific process. The gas flow is affected by the spinning process with increasing numbers of filaments concerning mass, drag and heat exchange.
The filament properties during the spinning process are characterized by observations from experiments. The filament position can be assumed to be a straight line between the individual capillary and the wind-up roller. Mass, momentum and heat exchange depend on the local filament properties: position, velocity, diameter, temperature, solvent concentration. For extended studies and other applications the spinning process could also be taken into account using fiber dynamics based on a one-dimensional Cosserat rod model along the fiber.
The evaporation is determined by the initial concentration in the spinning mass, the process conditions and the local concentration gradient between filament surface and surrounding gas. This model framework has been realized using Ansys Fluent as a standard tool for computational fluid dynamics. Mass, momentum and heat exchange due to the filaments are homogenized based on the principle action equals reaction, and have been considered as sources with so-called

Fig. 1
Spinning tower for melt-spinning process with angular shape (left) and dry-spinning process with rectangular shape (right) – black arrows indicate gas inlets and outlets

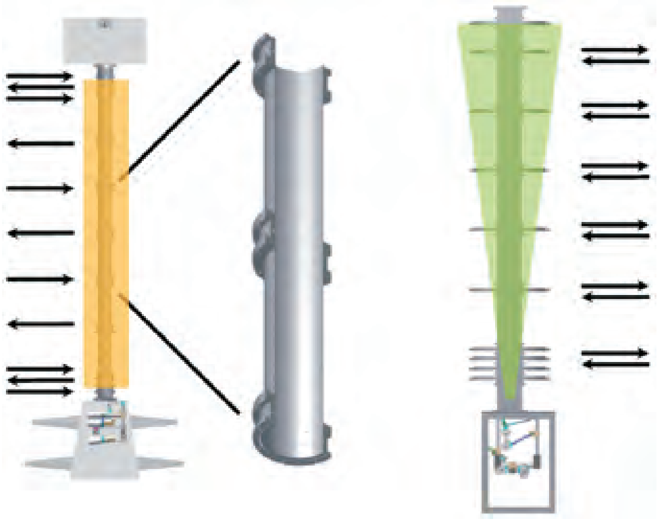
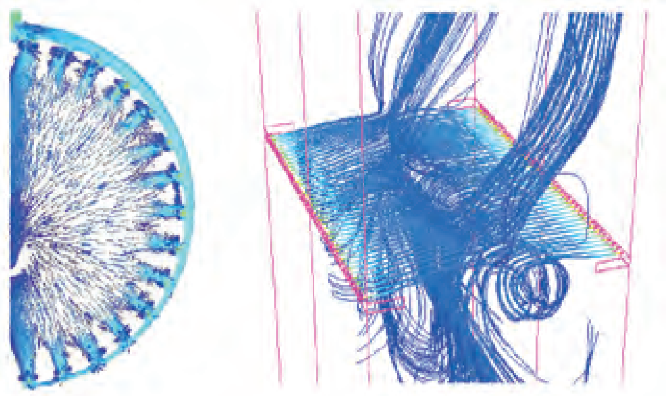


Fig. 2
Streamlines of optimized ring flange (symmetric half) (left) and rectangular flange with holes at the long sides (right)

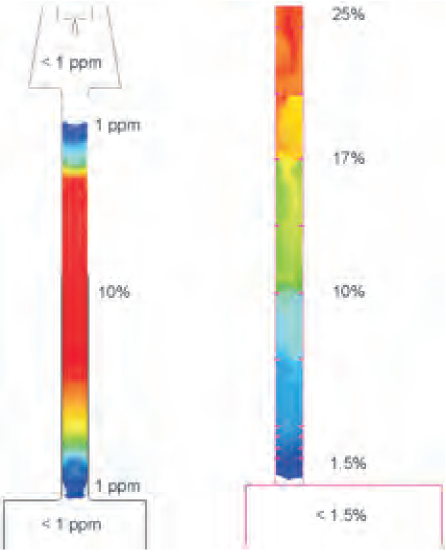


user-defined functions in Fluent. The model and simulation results have been validated in selected experimental settings. The main task is the usage of computations for the upscaling of the equipment and process conditions towards hundreds of filaments instead of tens.

Results and applications

The upscaling is demonstrated for the melt- and dry-spinning of Si-polymers. In the melt-spinning application, the filaments already have to be cured inside the spinning tower by using a curing gas. To generate a nearly homogeneous distribution inside the tower the inert gas is blown in and sucked out at several flanges with the option of adding curing gas (Fig. 1, left). For the upper glove box the most critical point is the towing effect with downward gas flow caused by the movement of the filaments and a corresponding back flow outside the filament bundle in upward direction. With respect to the one-sided inlet, it was analyzed with simulations that an axial-symmetric realization of the ring flanges will not lead to an equal outflow through all the holes. Therefore, it was necessary to

Fig. 3
Simulated gas flow under influence of spinning process with curing gas concentration in melt-spinning (left) and solvent concentration in dry-spinning (right)



adapt the ring flanges on basis of simulations.

The proposed solution keeps uniform holes and modifies the cross-section of the outer ring so that the outflow through all holes is nearly equal (Fig. 2, left). The second step is a local sequence of blowing-sucking-blowing at both ends of the spinning tower to avoid the appearance of curing gas in the glove boxes. Finally, the optimal process conditions are determined in a parametric simulation study which takes into account the mass flow at all inlets and outlets including additional curing gas together with flushing and suction in both glove boxes.

The simulation-based setting for the pilot plant scale with hundreds of filaments is shown in Fig. 3 on the left for the curing gas concentration: specified uniform concentration in the main part of the tube and no curing gas in the glove boxes (less than 1 ppm). This design of the spinning tower was realized and the process settings were validated with further improvements during the following experiments. Ceramic fibers with a filament number of up to 1,000 were produced on spools (Fig. 4).

Fig. 4
Realization of melt-spinning process and produced ceramic high-tech fibers

For the dry-spinning of a polymer solution, the task is mainly to realize a proposed solvent concentration gradient in the inert gas atmosphere which decreases. For low numbers of filaments additional solvent has to be blown in, whereas for large numbers the amount of solvent evaporating from the filaments is enough to generate a sufficient concentration. Its level is needed to control the viscosity along the filaments by the local solvent concentration inside the filaments. Due to a higher upscaling potential, a rectangular tube was preferred where the flanges act from both long sides. The streamlines of the simulation based design for these rectangular flanges are visualized in Fig. 2 on the right. The option of simultaneous blowing and suction on opposite sides has been rejected fully based on the simulation studies. The spinning tower includes only a lower glove box where a subcritical solvent concentration has to be reached due to safety requirements. Again a combined sequence of blowing and sucking is used above the lower glove box. The whole setting of flanges is shown in Fig. 1 on the right and the resulting optimal setting based on a parametric simulation study is shown in Fig. 3 on the right with respect to the solvent concentration. The maximum concentration on top is 25 % decreasing downwards with a concentration in the glove box under the specified limit of 1.5 %. The system was realized at Fraunhofer ISC with the aim to validate the simulations and to improve the process conditions for fiber spinning.

Conclusions

The usage of simulation models and computations in the upscaling procedure from laboratory to pilot scale was demonstrated to be extremely effective for 2 applications. The reason for that is the significant change of the gas flow from low numbers to large numbers of filaments due to size of the spinning tower, towing effects and evaporation of solvent which can be predicted by simulations. More sophisticated models can include a model of the filament spinning to improve the filament properties, the productivity and finally also to optimize the uniformity concerning all individual filaments. For both applications, a spinning tower was built according to the requirements derived from the simulation results. In both cases, by experimental studies, the concepts were proven to work. ■