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Stochastic 3D modeling of non-woven materials with wet-proofing agent

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ABSTRACT

A novel, realistic 3D model is developed describing the microstructure of non-woven GDL in PEMFC which consists of strongly curved and non-overlapping fibers. The model is constructed by a two-stage procedure. First we introduce a system of random fibers, where the locations of their midpoints are modeled by a 3D Poisson point process and the fibers themselves by random 3D polygonal tracks which represent single fibers in terms of multivariate time series. Secondly, we transform the random fiber system into a system of non-overlapping fibers using an iterative method leaned on the so-called force-biased algorithm. The model is validated by comparing transport-relevant characteristics computed for experimental 3D synchrotron data, and for realizations sampled from the stochastic microstructure model. Finally, we suggest a model for the spatial distribution of PTFE, a wet-proofing agent often used in non-woven GDL, and combine this PTFE model with our new microstructure model for non-woven GDL.

1. Introduction

Proton exchange membrane fuel cells (PEMFC) are a seminal technology for the generation of electrical power from hydrogen being efficient and eco-friendly. A key component of PEMFC is the gas-diffusion layer (GDL), whose main functions are the gas supply of the electrodes, the regulation of water storage, and evacuation within the GDL. The microstructure of non-woven GDL consists of highly curved fibers. Their morphology strongly determines the efficiency of the fuel cell as well as its stability [10,13,16]. Thus, the optimization of the GDL's microstructure is of utmost importance.

However, even for a known 3D microstructure of GDL material, it remains a challenging task to quantitatively predict its physical properties. In practice, a suitable microstructure for the GDL is usually determined by experiments based on trial-and-error. A more sophisticated way for the design of GDL microstructures is to generate synthetic ones using validated stochastic models. Then, by means of model-based computer experiments, microstructures can be detected such that the physical processes are improved. More precisely, by varying

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the values of the model parameters in a systematic manner, a large variety of different microstructures can be simulated. Subsequently, the goodness of these microstructures can be evaluated by numerical (transport) processes computed on the synthetic microstructures. Thereby, 3D microstructures with enhanced transport properties can be identified.

In this paper, we present a novel, realistic 3D model describing the microstructure of non-woven GDL in PEMFC which consist of strongly curved and non-overlapping fibers. There exist several stochastic models of fiber-based materials in literature, where in [2,4–6,12,19,21–23] models are considered which are designed for fiber-based materials consisting of straight fibers. In contrast to these approaches, in [1,9,18] stochastic models are proposed which describe materials consisting of curved fibers.

The modeling approach introduced in [1] is based on simulating a chain of spheres where the sphere midpoints follow a random walk. Thereby, each sphere chain is assigned a main direction according to a beta-orientation distribution. Given the preceding and the main direction of the sphere chain, the next direction is drawn from a multivariate von-Mises-Fisher distribution. In this way, curved fibers can be simulated using a small number of parameters. Moreover, this modeling approach offers a nice control of the directional distribution of the fibers. In particular, the main direction of a single fiber can be adjusted. However, when it comes to modeling fibers which exhibit extremely large curvature as loops, for example, other modeling approaches appear more suitable. In [9], a multi-layer model for curved, but horizontally orientated, overlapping fibers is introduced, where 2D random polygonal tracks representing planar fibers are simulated according to multivariate time series. Thereby, for instance, it is possible to simulate fibers exhibiting repeating loops, see also Fig. 1, by adjusting the parameters of the time series adequately. The benefit of the times-series approach is that the correlation of consecutive line segments can be taken into account. This is a great advantage for modeling the local course of fibers. Note, however, that the assumption of solely horizontally orientated fibers, is rather restrictive and not fulfilled for many fiber-based GDL materials.

In [18], another multi-layer model for systems of non-overlapping fibers is introduced. This modeling approach seems to be suitable to describe the structure of woven fiber materials. Note, however, that in the example of application considered in the present paper, we aim to describe non-woven GDL, which exhibit strongly curved fibers.

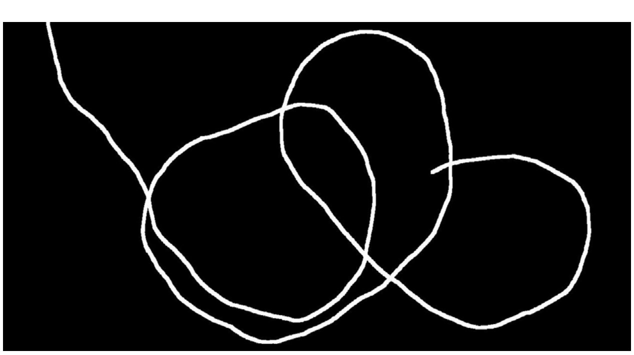

Fig. 1 – Planar fiber generated by the random polygonal track model based on multivariate time series.

In the present paper, we introduce a novel model that describes non-overlapping fibers with strong curvatures and apply this model to a non-woven GDL material consisting of carbon fibers. Thereby, a fiber model is used which is an alternative to the model proposed in [1]. In particular, the fibers drawn from our model can exhibit arbitrarily large curvatures (e.g. loops) in contrast to those obtained by the model introduced in [1]. But, on the other hand, the directional distribution of fibers obtained by our model can not be controlled in such an efficient way as in [1]. The fiber model considered in the present paper is a generalization of the multi-layer approach given in [9]. More precisely, fibers are now directly modeled in 3D (i.e., not by a multi-layer approach) using a single 3D time series. Furthermore, in the microstructure model proposed in the present paper, an avoidance algorithm is incorporated which prevents the fibers from mutual penetrations. Thus, the proposed model can describe fiber-based materials more realistically than the model considered in [9].

>In addition, our model allows the consideration of superstructures, e.g. clusters of fibers, which are observed in real non-woven GDL materials. Finally, since non-woven GDL are often treated with polytetrafluoroethylene (PTFE) (to increase hydrophobicity and to reduce blocked pathways caused by water holdups), we incorporate a wet-proofing agent (PTFE) into the model.

The stochastic microstructure model proposed in the present paper is constructed by means of methods from stochastic geometry and multivariate time series analysis (see e.g. [11,17,20]. and [7,15], respectively). In particular, a two-stage approach is used, where we first consider a germ-grain model to generate a system of overlapping fibers. The germs form a homogeneous Poisson point process in 3D and the grains are random 3D polygons described by a suitable multivariate time series, which are spherically dilated in 3D. Subsequently, the germ-grain model is transformed in the following way: First the fibers are translated such that they are evenly spread in space. Secondly, like in [1], an iterative avoidance algorithm is applied to the translated fiber system to eliminate overlaps between the fibers. This model is then used to suitably describe the microstructure of non-woven GDL in PEMFC. More precisely, the parameters of the non-woven GDL model are fitted to 3D image data of experimental non-woven GDL, where the image data is gained by synchrotron tomography. To adequately fit the model parameters, we extract a system of single fibers from experimental image data of non-woven GDL using an algorithm described in [8]. From the extracted fiber system, important properties like curvature measures or directional distributions of the fibers can be incorporated into the model. Finally, for model validation, transport-relevant characteristics computed for experimental data of non-woven GDL and those computed for virtual non-woven GDL drawn from the fitted model are compared to each other, where a good agreement is found.

The paper is organized as follows. Section 2 briefly describes an algorithm for the detection of single fibers in 3D synchrotron images. In Section 3, the stochastic model for 3D polygonal tracks is introduced. Section 4 establishes the 3D model for non-woven GDL. In Section 5 it is shown how PTFE can be included into the model as a further material component. Section 6 concludes the obtained results.