

Macromolecular Nanotechnology

Structurally and chemically heterogeneous nanofibrous nonwovens via electrospinning

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Abstract

Homogeneous nonwovens composed of polymer nanofibers of a given diameter are characterized by structural parameters such as the average pore sizes and internal surfaces as well as by transport properties, which are strongly correlated to the fiber diameter at a given porosity. Such nonwovens are used among others for filter applications, protective clothing or as scaffolds for tissue engineering. A frequent requirement is that, to be able to prepare nonwovens optimised for the specific application, one has to find ways to disrupt this strong correlation allowing independent modification of pore diameter, transport properties and internal surface or to induce local chemical and structural heterogeneities within the nonwoven. The route explored in this paper is based on the electrospinning of heterogeneous nonwovens composed of nanofibers with two different average diameters (by a ratio of up to 10 and more) on the one hand and/or different chemical nature on the other hand. Spinning parameters have been optimised to achieve this goal. In addition, nonwovens composed of fibers with circular cross-section and with ribbon-like cross-section have been prepared.

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1. Introduction

In electrospinning, a strong electrical field is applied to a droplet formed from a polymer solution or polymer melt at the tip of a die acting as one of the electrodes. The charging of the fluid leads to a conical deformation of the droplet and eventually to the ejection of a jet from the tip of the cone

[1–4]. The charged jet is accelerated towards the counter electrode, and thins rapidly during this period due to elongation and evaporation of the solvent until solid fibers are deposited onto the substrate located on the top of the counter electrode in general in a random fashion. The fiber deposition rate amounts to a several 10 m per second and the fibers are, in principle, infinitively long. The fiber formation can be controlled by a broad range of processing parameters, including viscosity, electric conductivity, surface tension, electric field strength and solvent concentration, for the diameter to be in the range from a few nanometers up to a few μm .

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The nonwovens resulting from electrospinning are, in general, characterized by a very high total porosity of up to 95% – i.e. a solid fiber fraction of only about 0.05 – and the fibers are usually randomly oriented in a plane defined by a planar counter electrode. Extended Molecular Simulations have been performed on nonwovens composed of micro- and nanofibers, which point out a strong correlation between fiber diameters and structural features of the nonwovens – such as the average pore diameter and the internal specific surface area – on one hand and between fiber diameters, transport properties – such as the diffusivity of gas or the permeation of gases and fluids – and corresponding pressure drops on the other hand [5–10]. Furthermore, filter collection efficiencies were evaluated and the total porosity turned out to be a main parameter. The effect of specific 1-d, 2-d and 3-d fiber orientations on the structure and the transport properties was investigated as well as the modifications in the transport properties as one goes from the ordinary gas transport regime via the transition regime to the Knudsen regime characterized by fiber and pore diameters smaller than the mean free path length of the gas molecules.

Nonwovens composed of polymer nanofibers are of interest for a broad range of applications. Highly effective filters including aerosol filters, protective clothing equipped with catalysts and protecting versus chemical and biological attacks, thermal insulation for textile applications but also scaffolds for stem cell growth and differentiation in the area of tissue engineering are examples for applications which benefit from the specific pore structure and the transport and surface properties controlled by the pore structure. Thermal insulation/wind resistance and tissue engineering may serve as an example [11–15]. Thermal insulation is primarily controlled in high porosity nonwovens by the diffusivity of the gases within the nonwoven. By reducing both the fiber diameter and the pore sizes in the nm range, one is able to approach the Knudsen transport regime, which causes a significant increase of the thermal insulation properties of the nonwoven well below the one of the pure gas. In the case of tissue engineering, extended studies have revealed that particular fiber diameters mimicking the ones found in the extracellular matrix in the body and the specific pore diameters which come with the fiber diameters in nonwovens strongly support stem cell proliferation and differentiation [13–15].

The rather strong correlations apparent from the simulations between fiber diameter and other struc-

ture and transport properties discussed above often make an optimisation of a nonwoven system for a particular application rather difficult. Techniques have been proposed and theoretically evaluated allowing the control of the total porosity in the nonwoven. Both the use of more flexible fibers such as elastomeric ones and the deposition of a shell layer on the fibers within the nonwoven, for instance, by chemical vapour deposition, can cause a significant increase of the fiber content within the nonwoven with corresponding modifications of pore diameter and transport properties [8,9]. The choice of flexible fibers, however, limits the range of applications of these nonwovens and the preparation of core-shell nanofibers as the basis of nonwovens strongly restricts the choice of the materials from which such a nonwoven system can be prepared. The preparation of structurally or/and chemically inhomogeneous nonwovens by electrospinning offers a promising route towards further optimisations in various applications, for instance, tissue engineering [16].

So far most investigations on fiber and nonwoven preparation via electrospinning aimed at creating nonwovens composed of uniform fiber diameters and fibers of one particular material as this facilitates the experimentally and theoretically based analysis of structure-property correlations. However, strong arguments can be put forward preparing nonwovens composed of well-defined fiber diameters which, however, differ either in size or in chemical composition. Tissue engineering and the flow of gases through the nonwoven may serve as examples again [16,17].

At a given porosity, the pressure drop in filters is controlled predominantly by the average pore size, whereas the collection efficiency is governed by the internal surface. As far as nonwovens with homogeneous fiber diameters are concerned both are associated with each other via the fiber diameter [5–10]. The total porosity can be considered as a fixed quantity since both simulations and experiments show that the total porosity of nonwovens obtained by deposition of solid fibers is in the range of 95%. Aiming at increasing the collection efficiency one should strongly reduce the fiber diameters. Yet this in turn increases the pressure drop. One way to avoid this problem is to prepare nonwovens of fibers which differ significantly in diameter. Thicker fibers can be used to control the average pore size and thus to keep the pressure drop low, whereas a small concentration of very thin fibers serves the purpose

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