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Effect of humidity on the generation and control of the morphology of honeycomb-like polymeric structures by electrospinning

Tian Liang, Maryam Parhizkar, Mohan Edirisinghe, Suntharavathanan Mahalingam *

Department of Mechanical Engineering, University College London, Torrington Place, London WC1E 7JE, UK

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ABSTRACT

Honeycomb-like 3D polymeric structures are useful to grow cells inside pores or in between nanofibres in cellular containment but their generation and morphology control involves different processing and forming procedures. In this work, using high speed camera footage, we show the dramatic effect of relative humidity (RH) has on the deposition of honeycomb structures using a hydrophilic water-soluble polymer. Approximately 35 mm diameter deposits were generated under several conditions of humidity while all other process parameters were kept constant. The morphology of the structures, i.e., their porosity and nanofibres, were studied and are related to the jet behaviour characteristics to explain why 3D honeycomb structures are obtained under selected optimum humidity conditions. 3D honeycomb structures were obtained only at an optimised 73% RH.

1. Introduction

Electrospinning enables generation of continuous polymer fibres by using a high electric field strength to overcome the surface tension of the polymer solution [1,2]. In the 1990s, scientists discovered organic polymers could generate nanosize fibres [3]. Since then, many investigations have been conducted to study the generation, structure and the properties of fine fibres [4–8]. In 2001, Deitzel et al. [9] observed that the residual charge on polymer solutions could be organised into a non-woven textile structure under certain processing conditions, they also observed the evolution of a honeycomb shape. A few years later, Thandavamoorthy et al. [10] discovered a self-assembled honeycomb shape of polyurethane nanofibres. They also found that the honeycomb structure formed well on a low conductivity substrate, and the accompanying nanofibres contained a number of beads. Yan et al. [11] generated self-assembled honeycomb-like structures by using polyacrylonitrile, polyvinyl alcohol, and polyethylene oxide individually. They demonstrated that solution properties and processing parameters affect the formation and assembly of honeycomb-like structures. Patra et al. [12] conducted a parametric study on manufacturing poly(lactic) acid nanofibrous mats by electrospinning and related their morphology to the production process. Recently, Ahirwal et al. [13] used both simulation and mechanical techniques to form hierarchical structures and explained that the repulsive electrostatic force of spun poly(ε-caprolactone) polymer helped to align them to form 3D porous structures. They also discovered that by increasing the spinning time the pores at the bottom of the structure were smaller than those in the higher layers.

Very recently, Liang et al. [14] using polyethylene oxide solution methodically investigated the effect of polymer concentration, nature of collection substrate, the working distance, and the formation time on the morphology of self-assembled honeycomb-like structures and nanofibres.

* Corresponding author. Tel.: +44 2076793920.
E-mail address: suntharavathanan.mahalingam@ucl.ac.uk (S. Mahalingam).

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However, most of the research published so far has only addressed the effect of solution properties and processing parameters on the formation of self-assembled honeycomb-like structures. The study of the effects of ambient parameters on the formation of honeycomb-like nanofibrous structures is still incomplete and needs systematic investigation. Hajra et al. [15] reported that higher temperature caused a better coalescence filtration performance on glass fibre medium, and the glass fibre medium assisted the organisation of structures well with higher relative humidity. Mit-uppatham et al. [16] have observed that the diameter of polyamide-6 fibres reduced by increasing the temperature of the polymer solution. It has been demonstrated that thinner poly(vinyl alcohol) nanofibres were obtained with the lower humidity and also the rate of evaporation is improved by decreasing the humidity [17]. Also, increasing the atmospheric temperature caused the diameter of cellulose acetate nanofibres to decrease, and higher humidity produced larger diameter nanofibres and increased the number of beads on the nanofibres [18]. A higher relative humidity can also enhance the breakage of poly(ethylene glycol) fibres and disturb their morphology [19]. The humidity also affected the pores generated on polycaprolactone fibres [19] and, in particular, raising the relative humidity decreased the quantity of poly(carbonate urethane) fibres collected [19].

In summary, many researchers have reported that ambient parameters can affect the morphology of fibres prepared from many different types of polymer [20–25]. However, there is no focussed research on the effect of ambient parameters on the formation and morphology of nanofibres and honeycomb-like structures which are important cellular reservoirs [14], especially there is no systematic study on the effect of changing humidity. Accordingly, the aim of this work is to uncover the relationship between the relative humidity and the generation of 3D self-assembled honeycomb-like polymeric structures and their nanofibres. In order to achieve this we have used a hydrophilic polymer (polyethylene oxide). Electrospinning and fibre deposition features were observed throughout using a high speed camera. Use of a hydrophobic polymer, ethyl cellulose, was also considered. However, especially at high humidities, stable fibre spinning over prolonged period was not possible to achieve reproducible trends.

2. Experimental details

2.1. Materials and solutions

Polyethylene oxide (PEO, molecular weight 200,000 g/mol, supplied by Sigma–Aldrich (UK)) was used to prepare the solutions. Distilled water was used as a solvent to dissolve PEO to make a polymer solution of 15 wt.% concentration. The PEO solutions were stirred for 12 h with a magnetic stirrer at the ambient temperature of ~25 °C and a relative humidity of 53%. A Testo 610 handy temperature/humidity metre (Testo Ltd., Alton, UK) was used to measure relative humidity and temperature. The physical properties of distilled water and PEO solutions were measured (Table 1), and included surface tension, density, viscosity and electrical conductivity. Distilled water was also used as a reference material throughout this investigation. The surface tension was measured by a Kruss Tensiometer K9 (Standard Wilhelmy's plate method). The viscosity was obtained by using a digital rotational viscometer (Brookfield, Harlow, UK). The electrical conductivity was measured by a conductivity probe (Jenway 3540, Bibby Scientific Ltd., Stone, UK). Each physical property of the PEO solutions and distilled water was measured five times and the mean value was determined. During repeated measurements, the equipment was cleaned with ethanol before each reading was taken to avoid errors.

Table 1
Physical properties of PEO solutions and distilled water used in this investigation.

Solution	Surface tension (mN/m)	Density (kg/m^3)	Viscosity (mPa s)	Electrical conductivity (mS/m)
Distilled water	72 ± 1.1	1026	1.3 ± 0.1	5.3 ± 0.1
15 wt.% PEO solution	71 ± 3.7	1053	2666 ± 53	25.0 ± 2

2.2. Electrospinning

Fig. 1a shows a schematic drawing of the experimental set-up of the electrospinning process. The stainless steel nozzle with an inner diameter 0.8 mm and an outer diameter of 1.1 mm was provided by Stainless Tube & Needles Co Ltd. (Tamworth, UK). One end of the nozzle was connected to a plastic syringe (Becton, Dickinson and Company, Oxford, UK) with silicone tubing (VWR International Ltd., Lutterworth, UK). The plastic syringe was clamped onto a syringe pump (Harvard Apparatus Ltd., Edenbridge, UK) to control the solution flow rate. The other end of the nozzle was connected to a high voltage power supply (Glassman Europe Ltd., Bramley, UK) and it was used to vary the electric field strength between the orifice of the nozzle and the collecting substrate. A stainless steel laboratory jack was placed underneath the nozzle and it was kept grounded. For each processing experiment, three glass microscope slides were aligned on the surface of the laboratory jack to collect the nanofibrous structures. The relative humidity surrounding the electrospinning devices were varied between 53% and 93% to investigate the effect on the nanofibres and structures produced by electrospinning. The ultrasonic humidifier CM50B (HoMedics Group Ltd., Tonbridge, UK) was connected to the insulated chamber using plastic tubing to control the degree of wetness within the chamber. One end of the tubing was fitted into the insulated chamber, the diameter of the fitting hole was equal to the outer diameter of the tubing. The other end was covered and sealed on the top of the mist outlet. The charged stainless steel nozzle and the laboratory jack were enclosed in an insulated perspex chamber. All the experiments on the generation of the nanofibres and structures were repeated three times to check the reproducibility of results.