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Electrospinning of Ethyl Cellulose Fibres with a Heated Needle and Heated Air Using a Co-axial Needle: a Comparison

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Abstract—**The concept of heated needle and heated air electrospinning process is presented. These techniques allow reduction in fibre diameters by increasing jet attenuation rate. Two experiments were carried out to see the effect of heat on ethyl cellulose fibre diameter. Different concentrations of ethyl cellulose in solution were prepared using a binary solvent system of ethanol and de-ionised water. The optimal concentration of ethyl cellulose was investigated along with parameters such as voltage and flow rate which can be used for electrospinning. The heating provided by the air stream via the co-axial needle generated thinner fibres (diameter up to < 1 μm), as compared to the heated needle electrospinning (diameter < 10 μm).**

Index Terms—**Co-axial, electrospinning, ethyl cellulose, heated needle, heated air.**

I. INTRODUCTION

Since its inception over a century ago [1],[2], electrospinning has gained much popularity only over the last two decades as researchers all over the world started to realize its immense potential in the field of nanoscience and nanotechnology [3]. This technique allows the production of fibres with diameters ranging from micrometres (e.g. 10-100 μm) to nanometres (e.g. 100 nm) from a vast range of materials such as polymers, composites, semiconductors and ceramics [4],[5]. Several beneficial characteristics appear at the nano-scale such as very large surface area to volume ratio, flexibility in surface functionalities, and superior mechanical properties (e.g. stiffness and tensile strength) [4]. Application of electrospun fibers include tissue scaffolds, protective clothing, filtration, nano-electronics [5] and food-grade films [6]. One of the main attractions of the electrospinning technique is its simplicity and inexpensive setup. At a laboratory level, a typical setup for electrospinning consists of a high voltage power supply (up to ~30 kV), a syringe pump, a flat tip needle of known internal diameter and a collector. The parameters which affect electrospinning have been described in various studies to date. These parameters include solution properties, process control parameters variables and the environment [3]. Solution properties include electrical conductivity, viscosity, surface tension, polymer molecular weight and concentration, and dielectric constant. Process control variables are flow rate, applied voltage, distance between needle tip and collector, type of needle, collector composition. Environmental settings include temperature, humidity and air velocity [7].

The present study is a continuation of our food engineering based research activity incorporating ethyl cellulose and investigates the effect of a heated needle and heated air on co-axial electrospinning of food compatible ethyl cellulose fibres [8]. The application of heat to the electrospinning process in these two ways could potentially give more control over fibre morphology as the fibre aspect ratio can be manipulated in addition to other variable parameters such as applied voltage and solution flow rate.

II. MATERIALS AND METHODS

A. Materials

The main materials in this study were: Ethyl cellulose (EC) (molecular weight ~ 22,800, Sigma-Aldrich, Poole, UK). Ethanol (General purpose research grade with 97.7% purity, viscosity 1.3 mPa s, BDH Laboratory Supplies, UK) and de-ionised water. To study the effect of heating the needle, a 686 μm (internal diameter) stainless steel needle was used whereas to study the effect of heated air, a coaxial needle was used with an inner needle of diameter of 686 μm and an outer diameter of 810 μm (Stanley Engineering, Birmingham, UK).

B. Preparation and Characterization of Solutions.

To electrospin ethyl cellulose fibres a binary solvent system of ethanol and water was developed with a ratio of 80:20, respectively. EC solutions of 5-35 wt. % were made for this study. The EC powder was dissolved in the binary solvent system after being stirred for 2 hours at the ambient temperature (26°C). The EC solutions were characterised by measuring their viscosity, surface tension and electrical conductivity (Table I). The equipment used for these measurements was calibrated against reference data. The viscosity of EC solutions was measured using a Brookfield DV III Ultra viscometer and RheoCalc V3 software. The electrical conductivity and surface tension of the EC solutions was measured using HANNA HI 8733 conductivity meter (CamLab Ltd, Cambridge, UK) and Wilhemy's plate method using a Kruss tensiometer K9, (Kruss Surface

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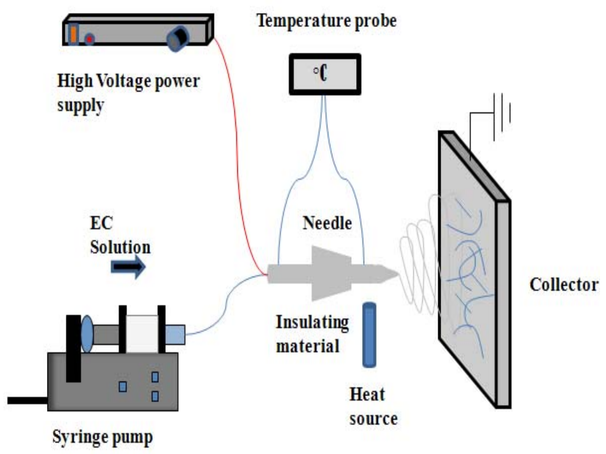
Science Centre, Bristol, UK) respectively.

C. Electrospinning Setup

The apparatus for electrospinning ethyl cellulose is illustrated in figure 1**.** The ethyl cellulose solution was infused through the needle using a Harvard Precision syringe pump (Model type PHD 4400, Edenbridge, UK). EC solution of 30 wt. % was found to be optimal to form fibres with minimal beads and hence used for this study. The flow rate of the solution was kept at 50 µl/min to obtain an adequate yield of fibres.

(a) Heated needle electrospinning of EC



(b) Co-axial electrospinning of EC with heated air

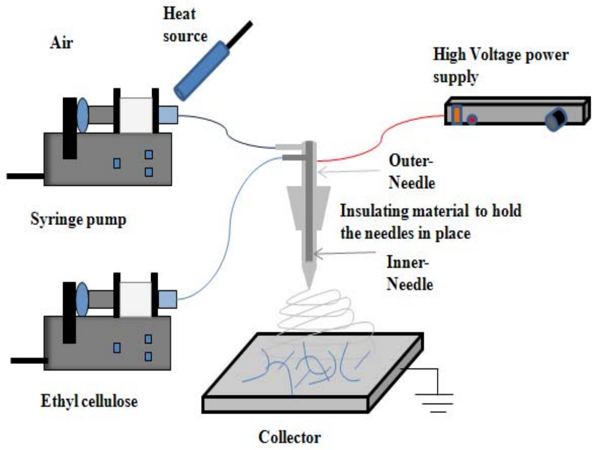


Fig. 1. Electrospinning setup used to see the effect of heated needle (a) and heated air (b) in this study.

The air fed into the outer needle of the co-axial needle setup was infused at a constant flow rate of 200 µl/min. The 30 wt. % EC solution was subjected to an applied voltage using a power source (Glassman High, Bramley, UK) attached directly to the stainless steel needle via a copper cable. The distance between the needle tip and grounded collector was set at 100 mm. The optimal value for applied voltage was found to be 16 kV in both cases by observing the jet formation using a high speed camera (Weinberger AG, Dietkon, Switzerland). The needle was heated via placing a Bunsen burner near the needle exit. The temperature was monitored by placing a probe on both ends of the needle. The air input was heated in a plastic syringe using a hot air gun (Steinel HG 2000 E, Sigma Aldrich, Poole, UK) (Fig.1). The rest of electrospinning setup for heated air experiment was the same as for the heated needle. The temperature in both experiments was carefully monitored by using the same temperature probe and all other processing parameters such as applied voltage and flow rate was kept the same to ensure experimental consistency.

D. Characterization of Electrospun Fibres

Electrospun fibres of EC were collected on glass slides and then observed under an optical microscope using a Nikon Eclipse (ME600, Nikon, Japan) camera at x5 magnification. The images were analysed using Acquis digital imaging software (Synoptics Ltd, Cambridge, UK). Further analysis of fibre morphology was performed using a Jeol JSM 6301F field emission scanning electron microscope (FE-SEM, JEOL Ltd, Herts, UK) was used. The fibres were gold sputter coated (Edwards Sputter coater S150B) for 3 minutes. An accelerating voltage of 5 kV was used with a working distance of 10 mm.

TABLE I: PHYSICAL PROPERTIES OF ETHYL CELLULOSE SOLUTIONS USED IN THIS STUDY.

Ethyl cellulose (wt.%)	Surface Tension (mN/m)	Viscosity (mPa s)	Electrical Conductivity (Sm^{-1} x 10^{-4}
5	25	23	47
10	29	100	61
15	36	363	68
20	54	1230	84
25	*	3692	800
30	*	11629	900
35	*	14600	1100

* Accurate surface tension measurements with the plate method not possible due to high viscosity of solution.

III. RESULTS AND DISCUSSION

The measured properties of the EC solutions are given in Table 1. The surface tension, viscosity and electrical conductivity of EC solutions increased with increasing polymer concentration. EC concentrations of 5-25 wt. % produced droplets and in some cases, short fibres with excessive beads. Fibres with minimal beads were obtained with 30 wt. %. (Fig. 2a). The 35 wt. % highly viscous EC did not result in any electrospinning due to the constant blockage of the needle. The effect of heat on the electrospinning of EC fibres was profound in both cases. A drastic reduction in fibre diameter was observed when the needle temperature was increased to 40°C (from ambient temperature ~26°C) (Fig. 2b) and then further narrowing was observed at 60°C (Fig. 2c). This can be attributed to the reduction of solution viscosity as heat is applied to the needle [9]. Increasing the temperature beyond 70°C resulted in very fast evaporation of ethanol which left a solid polymer residue in the needle. The fibre diameter obtained by this method of heating the EC solution decreased from 5 to 50 µm, with no heat, to 2 to10µm at temperatures in excess of 60°C.

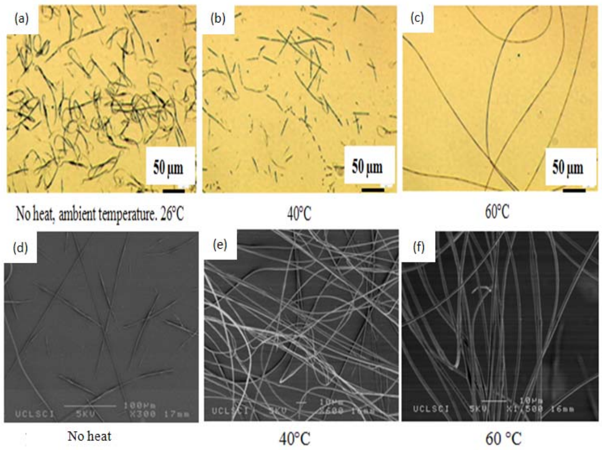


Fig. 2. Electrospun fibres obtained using 30 wt.% EC solution with a heated needle. Optical images (a to c) and scanning electron micrographs (d to f) of fibres with no heating of the needle, at 40 °C and 60°C, respectively.

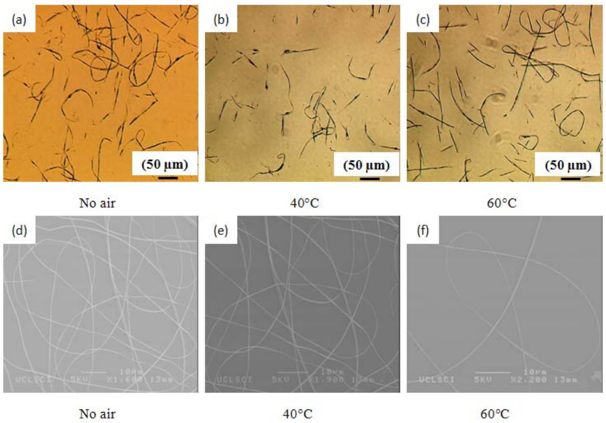


Fig. 3. Electrospun fibres using 30 wt.% EC solution with heated air. Optical images (a to c) and scanning electron micrographs (d to f) of fibres with no air (ambient temperature ~°26C), at 40°C and 60°C, respectively.

The effect of heated air blowing through the outer needle of the co-axial nozzle produced similar results. This configuration of combining the conventional electrospinning and a heated air stream device can delay the onset of polymer solidification due to evaporation of the solvent. The high flow rate of air can also provide additional drag force to the jet surface, leading to thinner fibres with a higher production rate [9]. Subsequently, due to additional deformation caused by the heated air stream, sub-micrometre scale fibres can be obtained from electrospinning. The drastic fibre size reduction can be seen in Fig. 3. The average fibre diameter decreased from 5 to 50 µm to <1 µm. Further work is being carried out to study the effect of temperature on solution properties and on optimisation of fibre aspect ratio (length and diameter) using both setups of electrospinning.

IV. CONCLUSION

The electrospinning of ethyl cellulose fibres via the two heating methods has been successfully used to produce sub-micrometre fibres. The fibres produced with heated air had smaller diameters, compared to the fibres produced via the heated needle for the equal temperature rise. With further process optimization, the diameters of the fibres could be made even smaller with the effect of the air becoming more pronounced and the applied voltage becoming a secondary deformation force in the electrospinning process.

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