Melt electrospun fibrous architectures with target geometries

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Abstract. In the melt electrospinning technique, the polymer melt is stretched under high voltage and the cooled to form microfibers structures with a fibre diameter in the tens of micrometres range, although some studies have reported values ranging from hundreds of nanometres to hundreds of micrometres. In this respect, this technique has significance in the biomedical field, where tissue engineering scaffolds with bimodal (nano and micro) fibrous structures are preferred in regard to cell adhesion, spreading and infiltration to final tissue reconstruction. This paper gives a review of recently reported melt electrospinning devices, especially those based on the direct writing principle, and of their comparison with the new melt Spraybase electrospinning device. The Spraybase device provides high precision melt jet deposition into 2D and 3D programmed architectures, with versatile translation speeds of the collector plate in the X-Y and the melt head in the Z direction. The melt spun fibrous architectures are designed depending on the types of tissue cells used in scaffold development.

1. Introduction
Electrospinning of polymer melt is a relatively new spinning technology, as seen by the publications in this field that all date after 2011. Literature on polymer melt electrospinning accounts for just 1% of the overall electrospinning literature [1]. Polymer melt electrospinning is an ideal technique for the production of highly porous nano- or microfiber structures that are suitable for biomedical use. In recent decades, melt electrospinning has been recognized as an ecological procedure, as it has eliminated the cytotoxic effects of solvents used in electrospinning [2]. As an ecologically acceptable method of spinning micro- and nanofibers, electrospinning has been attracting the interest of many scientists. There is increased interest also of direct 3D printing of biopolymers with a scaffold design in tissue engineering [3]. Melt electrospinning typically produces microfibers with a diameter ranging from 5–40 µm, though this technology is also capable of producing fibers with a diameter of just 0.5 micrometers [4]. In the field of regenerative medicine, this is highly applicable, as melt electrospinning is capable of producing tissues with a higher resolution than is possible using 3D printing [5]. In recent years, there has been a massive surge in the demand for polymer nanofibers, that are used for a variety of applications, including tissue engineering, protective clothing, filtration and sensor systems [1, 6].

2. Materials and methods
The polymer poly(lactic acid) (PLA) was used for sample preparation. This compound is obtained from natural resources and significantly reduces carbon traces in comparison with other oil-based plastics. The commercial name of PLA is Luminy® L175 (Figure 1). This is a homopolymer with a high melting
point and high viscosity that is suitable for fibre spinning. In comparison with standard PLA, this homopolymer polylactide has a higher melting point and accelerated crystallisation rate. The density of the polylactide is 1.24 g/cm³.

![Figure 1. Polylactide (PLA) [6]](image)

Polylactide electrospinning was performed on the device Spraybase® (AS-1204-000-01; Avectas Ltd., Ireland; Figure 2), at the Department of Fundamental Natural and Engineering Science, Faculty of Textile Technology, University of Zagreb. The polymer melt electrospinning device (Figure 2) consists of the following components: 1) high voltage source (voltage to 20 kV), 2) melt head with polymer container capable of movement along the Z axis, 3) heating system (achieves temperatures to 250°C), 4) air compressor for polymer emission (pressure to 5 bars), 5) metal flat plate collector capable of movement along the X and Y axes, with additional heating, and 6) safety cover.

![Figure 2. Polymer melt electrospinning device Spraybase®.](image)

Working with the polymer melt electrospinning device consists of several steps. The first step is development of the 2D model in the program “SEL program generator”. Next, the program and table of coordinates for collector movement is generated in the MSEL control system, which also controls the operation of the device. The desired parameters (voltage, pressure and temperature) are set on the device control panel.
Optimisation of process parameters pertains to air pressure, melt temperature, voltage and distance of the head from the collector [7]. Optimization involves setting the height of the head from the collector, speed of movement of the collector, number of electrospinning cycles (number of layers) and container filling with polymer granules.

3. Results

3.1. Sample porosity

Porosity was calculated for each sample from the first and second group. Porosity was calculated using a digital Dino Lite microscope to photograph every sample, and images were then processed using ImageJ software. After converting images into the appropriate 8-bit format (greyscale), a histogram was compiled, where 0 indicates pores and 255 indicates the polymer. The porosity results of individual samples are given in Table 1.

| Group 1 (single layer) | Group 2 (double layer) |
|-----------------------|------------------------|
| 1a                    | 0.70                   |
| 1b                    | 0.66                   |
| 1c                    | 0.68                   |
| 1d                    | 0.71                   |
| 1e                    | 0.74                   |
| 2a                    | 0.64                   |
| 2b                    | 0.56                   |
| 2c                    | 0.58                   |
| 2d                    | 0.62                   |
| 2e                    | 0.58                   |

The porosity results show a greater porosity in samples of Group 1 (1a), and therefore it can be concluded that adding a layer reduces the sample porosity (Figure 4).
The effective material constants for fiber network can be determined using homogenization procedure concept for fiber network. The strain energy fiber network for representative volume element is equal to strain energy continuum element with effective material constant \[\varepsilon_{ijkl} \]. The strain energy of the representative volume element under plane stress conditions are

\[
U = \frac{1}{2} \varepsilon_{ij} C_{ijkl} \varepsilon_{kl} V
\]

where \(V = b \cdot h \cdot 2 \cdot r\) - representative volume element, \(C_{ijkl}\) are effective elasticity tensor. We assume that microscopic deformation tensor of a fiber segments \(\varepsilon_{ij}\) is compatible with effective macroscopic strain \(\varepsilon_{ij}\) of effective continuum (affine transformation). This is bridge relations between fiber segment microstrain \(\varepsilon_{ij}\) and macroscopic strain \(\varepsilon_{ij}\) in the effective medium.

3.2. Air permeability

Air permeability was calculated in the group of samples with a single layer of PLA polymer created using polymer melt electrospinning. The samples of the first group, all with a single layer, called sample 1a, 1b and 1c were all spun under the same conditions though each sample is unique and therefore was tested separately. Cold air was blown through the sample (10x10 cm) at a rate of 1 m/s and permeability of each sample measured using an anemometer. Samples from Group 2, spun in a double layer, were measured in the same way. The results are given in Table 2.

| Sample (1 mm, single layer) | 1a   | 1b   | 1c   | 1d   | 1e   |
|-----------------------------|------|------|------|------|------|
| Thickness (mm)              | 0,4278 | 0,4080 | 0,6198 | 0,4360 | 0,4380 |
| Air permeability (m/s)      | 5,99  | 5,23  | 4,75  | 5,50  | 5,95  |
| Temperature (°C)            | 40    | 40.6  | 40.4  | 39    | 40    |

| Sample (1 mm, single layer) | 2a   | 2b   | 2c   | 2d   | 2e   |
|-----------------------------|------|------|------|------|------|
| Thickness (mm)              | 0,492 | 0,5956 | 0,7376 | 0,690  | 0,723 | 0,723 |
| Air permeability (m/s)      | 5,31  | 4,47  | 4,08  | 4,11  | 4,21  |
| Temperature (°C)            | 39.6  | 39    | 39    | 40    | 39    |

From the results shown in Table 2, Group 1 samples proved to have a higher air permeability, and therefore it can be concluded that by adding more layers, the air permeability is reduced (Figure 5).
Figure 5. Air permeability of samples

From the above Tables 1 and 2, the thickness of each individual sample can be examined, with air permeability and the temperature of air passing through the sample. The thinnest sample was sample 1a (single layer) (Figure 6).

Figure 6. Sample thickness

A steady state, laminar, incompressible model has been adopted for the flow regime inside our virtual media. Implemented in the Fluent code [18] is used to solve continuity and conservation of linear momentum in the absence of inertial effects

\[ \nabla \cdot \mathbf{v} = 0 \]  
\[ \nabla p = \mu \Delta^2 \cdot \mathbf{v} \]

(2)  
(3)
The grid size required to mesh the gap between two fibers around their crossover point is often too small.

4. Conclusion
The polymer melt electrospinning technique is becoming increasingly popular, with a number of advantages over polymer solvent electrospinning techniques: more ecologically acceptable production conditions, ability to produce materials with specific geometry, and microfiber production, which has wide-ranging applications in biomedicine. In this study, microfiber materials were produced from polymer (PLA) melt electrospinning with a description of the design of the 2D model to make three groups of materials with differing geometry and filament density. The primary aim of this study was to determine the influence of geometry and density of filaments on porosity and permeability. By testing PLA material porosity and permeability, the following was established: 1) that increasing distance between filaments reduces the maximum force, corresponding strain at the same force and tensile strength, i.e. that a higher density of filaments gives higher tensile strength, but also deformation; 2) an analysis of the data in Tables 1 and 2 shows that sample 1a was the thinnest of the samples, in a single layer; 3) analysis of the porosity of samples showed that sample 2a in a double layer was the less porosity air permeability of the samples.

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