



# The effect of poly (lactic-co-glycolic) acid composition on the mechanical properties of electrospun fibrous mats



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## ABSTRACT

The aim of this study was to investigate the influence of polymer molecular structure on the electrospinnability and mechanical properties of electrospun fibrous mats (EFMs). Polymers with similar molecular weight but different composition ratios (lactic acid (LA) and glycolic acid (GA)) were dissolved in binary mixtures of *N,N*-dimethylformamide (DMF) and tetrahydrofuran (THF). The intrinsic viscosity and rheological properties of polymer solutions were investigated prior to electrospinning. The morphology and mechanical properties of the resulting EFMs were characterized by scanning electron microscope (SEM) and dynamic mechanical analysis (DMA). Sufficiently high inter-molecular interactions were found to be a prerequisite to ensure the formation of fibers in the electrospinning process, regardless the polymer composition. The higher the amount of GA in the polymer composition, the more ordered and entangled molecules were formed after electrospinning from the solution in THF-DMF, which resulted in higher Young's modulus and tensile strength of the EFMs. In conclusion, this study shows that the mechanical properties of EFMs, which depend on the polymer molecule-solvent affinity, can be predicted by the inter-molecular interactions in the starting polymer solutions and over the drying process of electrospinning.

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## 1. Introduction

Electrospinning of micro- and nanoscale fibers is a well-documented method for preparing scaffolds in tissue engineering (Yang et al., 2013; Francis et al., 2010; Dhandayuthapani et al., 2011; Jose et al., 2009; Szentivanyi et al., 2011) and for wound dressings (Boateng and Catanzano, 2015; Rieger et al., 2013). Three dimensional structures with high porosity can accommodate cell proliferation and differentiation to form extracellular matrix (Rieger et al., 2013). For successful applications, the physical structure should support the growth of cells and also withstand the compression and constriction *in vivo* (Tan and Lim, 2006; Prabakaran et al., 2012). In addition, the mechanical properties of an engineered scaffold should be comparable with the native tissue. Both natural and synthetic polymers have been electrospun

to produce such structures. The most extensively studied polymers include poly (lactic acid) (PLA), poly (glycolic acid) (PGA) and their copolymer poly (lactic-co-glycolic acid) (PLGA) (Singh et al., 2014). They are all biodegradable and biocompatible and are considered safe materials that are approved by the FDA for numerous applications in tissue engineering and drug delivery.

Several properties of the electrospinning feed solution, such as the viscosity, surface tension, conductivity, and evaporation rate, play an important role in the electrospinning process for producing satisfactory micro- and nanoscale fibrous structures. The surface tension, evaporation rate and electrical conductivity of the polymer solutions are often determined by the solvents used (Yao et al., 2008; Liu et al., 2017; Bock et al., 2012). It has also been reported that rheological properties of a polymer solution play a critical role in the process of fiber formation (Klossner et al., 2008; McKee et al., 2004; Kong and Ziegler, 2012). Molecular entanglement in the polymer solution is a prerequisite for fiber formation with electrospinning (Kong and Ziegler, 2012; Bhardwaj and Kundu, 2010; Shenoy et al., 2005). The existence of molecular

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interactions can be detected from the rheological properties of solutions. Literature on rheological behavior of polymer solutions at low and high concentrations suggest very clearly that two fundamentally different types of intermolecular interactions need attention. Firstly, the rheological behavior of a dilute polymer solution resembles a suspension consisting of discrete rigid particles (Batchelor and Green, 1972; Simha and Somcynsky, 1965; Graessley, 1974). The intermolecular interactions may be caused by the friction between discrete rigid particles (no overlapping between molecular chains) under fluid flow. As a result, the intermolecular interactions are not sufficient to resist the deformation caused by surface tension, and thus particles are formed instead of fibers (Christanti and Walker, 2001; Bohr et al., 2014). A second type of intermolecular interaction, the segment–segment contacts between molecules has been evaluated in concentrated solutions and melts where molecular chain-overlapping and even entangled networks form, which can resist the deformation caused by the flow fields of fluids. Besides the concentration, the solvent properties determine the molecular-level behavior in the solution (Gandhi and Williams, 1971; Gandhi and Williams, 1972; Servick, 2006). Different macromolecules have different affinity for the same solvent, which further affects its flexibility and inter-/intra molecular interactions. Moreover, the polymers that are more flexible and extended in the solution, exhibit higher density and can form molecular entanglements at lower concentration.

PLA, PGA, and their copolymer PLGA with different molar ratios of lactic to glycolic acid have different physicochemical properties, such as the hydrophilicity and solid state properties. Lactic acid (LA) is more hydrophobic than glycolic acid (GA), due to the existence of methyl groups. Therefore, the copolymer rich in lactic acid is more hydrophobic than the one with less lactic acid. When dissolved in the same solvent, copolymers of different LA-GA ratios have different affinity to the solvent molecules, and further, they influence the interactions between the polymer chains. The viscosity of the copolymer solutions will be affected accordingly. Moreover, PLA in its levorotatory (L-PLA) or dextrorotatory (D-PLA) form is semi-crystalline, while the racemic form is amorphous, and PGA is highly crystalline. It can be expected that the mechanical properties of the copolymers change as the composition is changed (D'Avila Carvalho Erbetta, 2012).

In order to investigate the influence of polymer composition on the electrospinning process as well as the mechanical properties of the resulting EFMs, PLA and PLGA with different molar ratios of lactic and glycolic acid were employed in this study. The molecular behavior of the polymer solutions were studied by a viscometer and a rheometer. The mechanical properties of the final EFMs were characterized with respect to the tensile strength and Young's modulus.

## 2. Materials and methods

### 2.1. Materials

PLA (R205, inherent viscosity (IV) 0.55–0.75dl/g), PLGA 7525 (RG 755, LA: GA 75:25, IV 0.50–0.70dl/g) and PLGA5050 (RG 505, LA:GA 50:50, IV 0.61–0.74dl/g) were purchased from Evonik (Darmstadt, Germany). Molecular biology grade of *N,N*-dimethylformamide (DMF), and HPLC grade of tetrahydrofuran (THF) were obtained from Sigma-Aldrich (Copenhagen, Denmark). All materials were used as obtained.

### 2.2. Measurement of molecular weight

In order to measure the molecular weight of the starting polymers used in this study the polymer samples were dissolved in chloroform at a concentration of 3–4 mg/mL, filtered, and analyzed via a size exclusion chromatography (SEC, SIL-10AD, Shimadzu, Kyoto, Japan). The sample solution (100  $\mu$ L) was injected into the system, consisting of PL-gel MIXED-D columns (300  $\times$  7.5 mm; bead diameter 5 mm) from Polymer Laboratories (Shrewsbury, UK) and the signals were detected using a differential refractive index detector (Viscotek model 200, Houston, TX, USA). The elution was performed at 25  $^{\circ}$ C with chloroform at a flow rate of 1 mL/min. Poly (methyl methacrylate) polymers with known molar masses were employed as standards for the calibration.

### 2.3. Measurement of intrinsic viscosity

Intrinsic viscosity of the starting polymer solutions was measured using an Ubbelohde Semi-Micro dilution viscometer (No. 50, N213, Cannon instrument Company, USA) at 25  $\pm$  0.2  $^{\circ}$ C, as

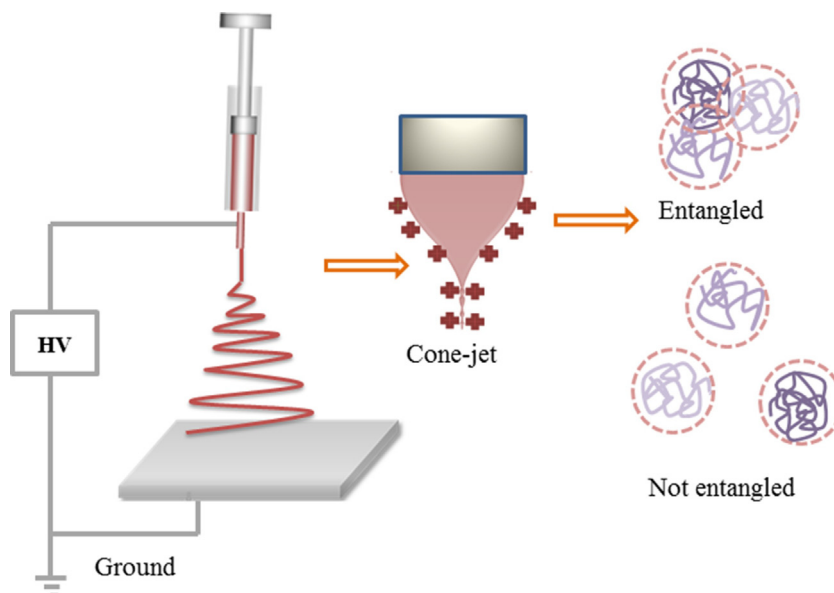


Fig. 1. Schematic illustration of electrospinning setup and the polymer molecular conformation in solutions.

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