Journal of the Mechanical Behavior of Biomedical Materials 72 (2017) 6–13

Contents lists available at ScienceDirect

Journal of the Mechanical Behavior of Biomedical Materials

journal homepage: www.elsevier.com/locate/jmbbm

Elastic moduli of electrospun mats: Importance of fiber curvature and specimen dimensions

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ARTICLE INFO

Keywords:
Fiber curvature
Electrospun
Specimen dimension
Tissue

ABSTRACT

Success of tissue engineering relies on the architecture and properties of porous scaffolds. Electrospun nonwoven scaffolds in the form of mats are unique materials due to large surface area to volume ratio, high porosity, versatility in surface functionalities and excellent mechanical properties. Maneuvering the mechanical behavior of the electrospun mat is a major challenge both from theoretical and experimental perspectives. Herein, we report a two-dimensional (2D) analytical model of normalized elastic moduli of electrospun mats by formulating a relationship with the governing fiber and structural parameters. The analytical model of normalized mat modulus has also accounted for fiber curvature in the form of sinusoidal curve along with the specimen dimensions considered during the uniaxial tensile test. A comparison has been made between the magnitudes of normalized mat modulus obtained through predictive modeling and the experimental results adapted from the literature. In general, a good agreement has been found between the theoretical and the experimental results of normalized moduli of electrospun mats. An interplay of some of the governing parameters has been analyzed through parametric analysis. Through theoretical modeling, the normalized amplitude of fiber crimp via fiber diameter along with the aspect ratio of specimen dimensions are observed to be the dominant factors responsible for modulating the normalized mat modulus.

1. Introduction

Over the last few decades, tissue engineering has emerged as a new paradigm for reconstructing damaged tissues and organs. Tissue engineering utilizes three basic tools, i.e. cells, scaffolds, and growth factor (Ikada, 2006). With the aid of cells and growth factor, the new tissues are being regenerated. The scaffold provides the suitable environment for these cells during the regeneration of tissues (Baldino et al., 2015). Specifically, the scaffolds should have an interconnected porous structure in order to ensure cell penetration along with the sufficient supply of nutrients to cells (O'brien, 2011). Simultaneously, the mechanical properties of scaffolds should match with that of anatomical site, and it should sustain the stresses during surgical handling. Electrospun scaffolds in the form of mats exhibit high surface area-to-volume ratio, high porosity, versatility in surface functionalities, and excellent mechanical properties (Huang et al., 2003). Superior mechanical properties of electrospun mats are pre-requisite to design and develop scaffold materials for tissue engineering. Modulating the mechanical behavior of the electrospun mat is a challenging task both from theoretical and experimental perspectives. Typically, nanofibers in the electrospun mat are fabricated keeping in view of their extraordinary mechanical properties in comparison to bulk materials (Tan and Lim, 2006). However, the characterization systems for experimentally determining the tensile properties of ultra-fine fibers are not easily available (Baji et al., 2010). The problem becomes further complicated when the electrospun mat consists of nanofibers with distribution of diameters (Richard-Lacroix and Pellerin, 2013). In the past, the elastic moduli of nanofibers were significantly enhanced when the diameter of nanofibers was kept below a critical level (Arinstein et al., 2007). Alternatively, the mean elastic modulus of the nanofibers can be predicted by formulating a relationship with the elastic modulus of the electrospun mat keeping in view that the tensile properties of mats can be easily determined (Wan et al., 2015).

Several analytical (Pai et al., 2011; Sun et al., 2008; Wan et al., 2015), semi-analytical (Ban et al., 2016; Donius et al., 2013; Rizvi et al., 2012; Rizvi and Pal, 2014), and numerical (Broedersz et al., 2011; Silberstein et al., 2012; Stylianopoulos et al., 2008; Wei et al., 2009) approaches have been employed to predict the tensile properties of electrospun mats. Majority of these studies highlighted a pitfall *whilst* a relationship has been formed between the tensile properties of electro-

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http://dx.doi.org/10.1016/j.jmbbm.2017.04.013
Received 28 February 2017; Received in revised form 7 April 2017; Accepted 10 April 2017
Available online 12 April 2017

Nomenclature

α	In-plane fiber orientation angle of the electrospun mat
β	Angle formed by the diagonal of the rectangular specimen with the test direction
γ	Normalized mat modulus of the electrospun mat
$\delta(\alpha)$	Deformation in the fiber oriented at an angle (α) due to C_{jp}
$\delta_p(\alpha)$	Component of $\delta(\alpha)$ in tangential direction
$\Delta(\alpha)$	Perpendicular distance between the axis of parallel fibers oriented at an angle (α) in a layer
$\Delta_w(\alpha)$	Projection of $\Delta(\alpha)$ in widthwise direction of the specimen
ε	Strain experienced by the electrospun mat
λ	Wavelength of fiber crimp
$\bar{\lambda}_p$	Statistical mean projection of λ on the stress direction
κ	Constant related to fiber crimp parameters
τ	Coefficient of variation of fiber diameter
T_s	Tensile stress applied to the electrospun mat
σ	Standard deviation of wrapped normal distribution
$\Omega(\alpha)$	Fiber orientation distribution function
a_0	Amplitude of the fiber crimp
A_f	Cross-sectional area of the fiber
B_1, B_2	Integrals related to bending and tensile strain of the fiber, respectively
ζ	Fiber crimp
C_j	Mean force experienced by the fiber in the electrospun mat
C_{jn}, C_{jp}	Normal and tangential components of C_j, respectively
d_f	Fiber diameter
$\bar{d}_f$	Mean fiber diameter
D	Diameter of the unit cell
E_f	Young's modulus of the constituent fiber
E_{mat}	Young's modulus of the electrospun mat
H	Integral used for calculating proportion of fibers contributing to the tensile resistance
I_f	Area moment of inertia of fiber
$J(\alpha)$	Effective width of the specimen for the fibers oriented at angle (α) that contribute towards the tensile stresses
$\bar{l}$	Mean length of fiber in the unit cell
l_λ	Undulated length of constituent fiber in one sinusoidal repeat
$L(\alpha)$	Total length of constituent fibers oriented at an angle (α) in the unit cell
L	Total length of constituent fibers in the electrospun mat
L_s	Length of the rectangular specimen
$n(\alpha)$	Number of constituent fibers oriented at an angle (α) in the unit cell
$n_j(\alpha)$	Actual number of constituent fibers orientated at an angle (α) in the defined specimen dimensions
N_j	Total number of constituent fibers contributing towards tensile resistance in the defined specimen dimensions
N_{ja}	Total number of fibers per unit area contributing towards tensile resistance in the defined specimen dimensions
q	Integral used for the averaging of crimp wavelength
p	Integral used for averaging parameter of force and deformation simultaneously
s	Aspect ratio of the specimen during the uniaxial tensile test
t	Thickness of the electrospun mat
V_f	Fiber volume fraction
W_s	Width of the rectangular specimen

spun mat and that of constituent fibers: how *realistically* the micromechanics of fiber mat has been captured via unit cell approach? Ideally, the unit cell approach should incorporate fiber orientation, mat porosity, fiber curvature, fiber-fiber bond characteristics, and entanglements (Baji et al., 2010). In addition, the theory should integrate the electrospun mat specimen dimensions, which can be used *whilst* validating with the experimental work. Some of the governing issues have been dealt sporadically, for example, Pai et al. (2011) have accounted for the fiber curvature while formulating a quantitative relationship between the elastic moduli of the electrospun mats and that of constituent fibers. On the other hand, a predictive model revealing the relationship between the tensile strength of a randomly oriented nanofiber mat, porosity, test specimen dimensions (width to length ratio), and the tensile strength of a single nanofiber has been proposed (Wan et al., 2015). However, a comprehensive analytical model of elastic moduli of electrospun mats based upon specimen dimensions, governing fiber and structural parameters has not been developed yet. Therefore, the principal goal of the research work was to develop a two-dimensional (2D) analytical model to formulate a relationship between the elastic properties of the electrospun mat and those of constituent fibers along with key structural characteristics of fiber mat. This comprehensive predictive model has also incorporated the specimen dimensions used for tensile test through a multistage modeling process. Initially, the structural parameters of an electrospun mat in a defined unit cell were computed. Subsequently, the proportion of fibers contributing to the tensile stresses in a defined specimen dimensions was predicted. Lastly, the elastic properties of electrospun mat and that of constituent fibers were mathematically related by introducing the fiber curvature in the form of sinusoidal path. The proposed model was also validated with the experimental data of Stylianopoulos et al. (2008) and Silberstein et al. (2012). Further, the parametric analysis was performed to elucidate the influence of specimen dimensions along with the governing fiber and structural parameters.

2. Theoretical analysis

Electrospun mats have similar structural characteristics as that of spunbonded nonwoven materials, which consist of continuous long lengths of stochastically aligned filaments. In the past, a circle of unit diameter was considered as a unit cell for predicting the mechanical properties of spunbonded nonwoven materials (Lee and Argon, 1983a). Accordingly, a unit cell of circular geometry consisting of electrospun mat has been considered in this analysis. In general, it is invariably assumed that the fibers are uniformly distributed within the unit cell, and the interactions between them have been neglected similar to the earlier works (Bais-Singh and Goswami, 1995; Cox, 1952; Wan et al., 2015). In addition, the following assumptions have been made in order to simplify the predictive model.

- Electrospun mat consists of continuous fibers having infinite length such that the ends of the fibers are gripped in the jaws during the tensile test. Fibers that are not gripped in jaws impart negligible contribution towards the tensile resistance (Wan et al., 2015).
- The distribution of constituent fibers is considered to be aligned in the in-plane direction such that a homogenous array of fibers is assumed in a 2D plane. The orientation of the fiber is defined by an angle (α) formed between the tangent to the fiber curvature and one of the principal in-plane test directions (Komori and Makishima, 1977).
- Constituent fibers of the electrospun mat have the alignment of these fibers that follow an orientation distribution function, $\Omega(\alpha)$, such that $\int_{-\pi/2}^{\pi/2}\Omega(\alpha)d\alpha = 1$.
- Shear stresses that are generated between the constituent fibers of the electrospun mat during the tensile test have been neglected.