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Finite element modeling of electrospun nanofibre mesh using microstructure architecture analysis

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This investigation is aimed at modeling the tensile behavior of electrospun polyurethane (PU) membrane. The PU web is produced with different morphologies and the structural parameters are studied through SEM images. Three-dimensional network is simulated using ABAQUS software. Each fibre is modeled as hyperelastic material and each crosslink is modeled as multi point constrain tie. The stress-strain behavior of PU mat is modeled by finite element method, and the effect of fibre diameter, fibre orientation and thickness of web is investigated. The stress–strain curves of networks at three different morphologies are compared with modeling measurements. The model by using third order reduced polynominal as fibre hyperelastic potential energy function shows good agreement with experimental findings which confirm that the tensile behavior of PU web can be explained entirely by microstructure of the network.

Keywords: Finite element modeling, Micro architecture, Nanofibrous membrane, Polyurethane, Protective clothing, Tissue engineering

1 Introduction

Electrospinning procedure has emerged as a promising method for fabricating fibrous materials in different engineering fields such as tissue engineering, filtration, energy harvesting, membranes, military and protective clothing applications. Electrospinning involves the use of a high voltage power supply to eject a polymer fibre from solution and deposit it onto a grounded target. Fibre diameter can be varied from 50 nm to more than 5 µm by controlling the electrospinning conditions, and orientation of the resultant network can be imparted by depositing this fibre onto a moving target[1,2].

Polyurethane (PU) has received considerable attention in electrospinning process due to easy care, resistance to microorganism, and excellent hydrolytic stability[3-7]. The physical and mechanical properties of the nonwovens are closely related to the geometric arrangement and fibre diameter[1]. Lee *et al.*[3] reported that electrospun PU fibre mats show non-linear elastic and inelastic behavior. In another study, Lee *et al.*[4] investigated mechanical behavior of electrospun fibre mats of PVC/PU polyblends. They reported that a large number of point bonded structures are presented in pure PU nanofibre mat and a linear elastic behavior up to fracture accrued in them. They noted that probably the reason for such thermoplastic elastomeric behavior is the formation of a large number of point-bonded structures between fibres in elastomeric PU that makes them a quasi-continues phase.

Tensile tests of hard materials within the elastic range give a stress–strain relationship in the form of a straight line of slope E, the elastic or Young's modulus. The modulus E is widely used to characterize the stiffness of hard materials. On the contrary, the tensile load–deformation relationship of elastomers is considerably nonlinear. The local slope of the load–deformation curve drops off significantly and the gradient of the curve changes to the extent the elastomer is stretched. Then the typical result of tensile test of elastomers is a stress–strain curve of S-shape, for which Hooke's law is not valid. Stress at different strain[8] and potential energy function are commonly used for the characterization of elastomer properties.

The macromechanical behavior of fibrous materials is sensitive to the microstructure architecture[9]. Stylianopoulos *et al.*[10] predicted the effect of fibre diameter and fibre orientation on tensile properties of

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electrospun fibre mesh using computational modeling. They determined the elastic modulus of fibre (E_f), by fitting the model to the experimental stress-strain response of electrospun mesh. PU fibre modulus of 18 MPa could fit this strain-stress model. They showed that the computed moduli of the meshes are lower than that of individual fibre (E_f), because of volume fraction and network effect. They assumed that microarchitecture (e.g. fibre diameter, orientation, angular standard deviation) is uniform throughout the mesh thickness, but spatial homogeneity of mesh architecture is not verified.

The utility of electrospun fused fibre meshes, however, is limited due to the absence of a theoretical framework to predict how the bulk mechanical properties depend on the properties of individual fibres. Such a framework is inherently multiscale, with the web dimension being on the centimeter length scale, while the underlying fibrillar architecture on the micrometer scale. The strong dependence of native nanofibrous structure response on fibre orientation, diameter, and reorientation in response to strain, suggests that it is necessary to consider the both length scales. Thus, the continuum constitutive models often cannot predict the tissue response under every loading condition. The scale separation makes multiscale, structure-based mathematical models as an attractive option to describe the mechanical behavior of nanofibrous structure.

In this study, nanofibre networks have been electrospun from a polyurethane elastomer, and fibre diameter and orientation are systematically varied. Mechanical testing is performed to determine experimentally the dependence of the tensile and elongation on the network microstructure. A structure model is also developed wherein homogenization techniques are used to study mechanical behavior of polyurethane. In this model, three dimensionality and interaction between the PU fibres are considered. The three-dimensional finite element method is employed for the analysis. Finally, the finding of model is compared with experimental findings.

2 Materials and Methods

2.1 Electrospinning Process

Electrospinning solution was prepared by dissolving 13, 14 and 15 % (wt./vol) commercial PU (Mw: 65,000, Bayer, Germany) in tetrahydrofuran (THF)/ N,N-dimethylformamide (DMF) mixture [60:40, 55:45, 50:50 (v/v)]. The efficiency of mesh thickness was obtained by performing electrospinning process for different durations (2,4, and 6 h). In the electrospinning process, the prepared solution was electrospun simultaneously on the rotating drum from two opposite nozzles (Fig.1). The distance between nozzles and collector was determined to be 130 mm. To separate nanofibres from the solution in the electrostatic field, the selected rotation speed and traverse speed of drum were 150 rpm and 400 mm/min respectively. Subsequently, a voltage of 13kV was applied to draw the nanofibres from the solution in the electrostatic field.

Figure 2 shows a typical SEM photograph of electrospun PU web. These pictures show that the fibres oriented in different directions and it is clear from the cross-section that there are lots of layers in one electrospun nanofibre mat.

Stylianopoulos *et al.*[9] used the tensile modulus of the bulk PU for nanofibre modulus. In this study, the aligned yarn is produced from the same solution of PU in the electrospinnig set up, as used by Najafi *et al.*[11]. Two nozzles with opposite charge were used to produce aligned nanofibres in yarn formation zone. These nozzles were connected to 1 mL syringes that were fixed on the syringe pump. Polymer solution was electrospun, produced jets were moving towards each other, and finally these jet joined together on the surface of aluminum collector. The aluminum drum was used as a temporary collector. Consequently, twisting and winding units were used to align and twist the nanofibre strand. During this transformation, a tensile load was exerted on the nanofibres and a spinning triangle was formed.

2.2 Characterization and Mechanical Testing

The thickness of membranes was measured using a micrometer (Dial Thickness Gage, Mitotoyo, Japan). The average of 20 measurement points was reported as the mat thickness. A scanning electron microscope (SEM) was applied to examine the morphology of

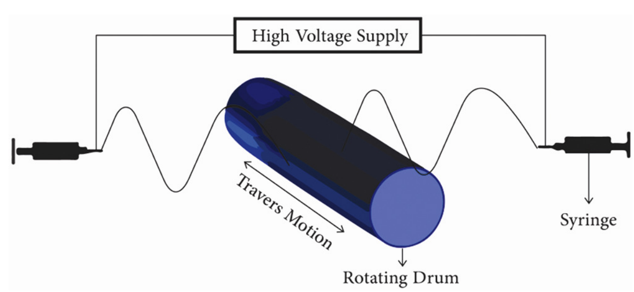

Fig.1—Schematic diagram of double nozzle electrospinning machine with rotating drum[1]

electrospun PU mats (440i, LEO Electron Microscopy, England). The diameters of the nanofibres were determined from SEM images using Image analyzer software[12]. The nanofibres length and orientation distribution were calculated by a program which was encoded in MATLAB software (version 7.0.0.1920)[1]. One hundred pieces of fibres in each sample were measured for determining the fibres orientation distribution. The sum of fibres orientation frequency at each 30^o interval was derived (Table 1). For characterization of tensile properties of the mats, dumbbell-shaped specimens were prepared and tested with a crosshead speed of 50 mm/min based on ASTM D-638. This test was carried out on five specimens in machine direction (MD) and five specimens in cross direction (CD). SPSS package 13 was used for statistical analyses.

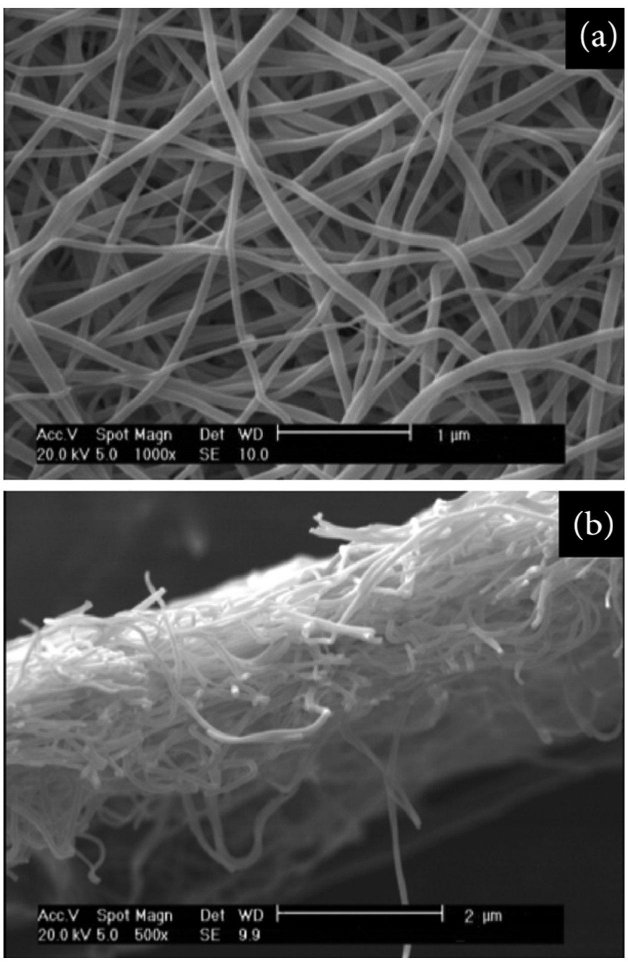

Fig. 2—Typical SEM photographs of electrospuns PU web (a), and cross-section (b)

Table 1—Degree intervals and corresponding mean length

Intervals, deg	0-30	30-60	60-90	90-120	120-150	150-180
Fibre length, µm	343.171	110.120	112.372	138.063	98.364	367.115

2.3 Modeling

The 3D network used in this study for modeling fibre mesh has the following assumptions:

- The fibre mesh structure is statistically homogenous
- The fibres are straight and oriented in the same plane (planar fibre mesh)
- The fibres are well distributed in surface, in the sense that they do not form bundles of parallel fibres
- The fibre diameters are uniform along the fibre length
- The deformation is nonlinear elastic
- All fibre contacts points are rigid

2.3.1 Representation of Fibre Network

The average values of fibres diameter, fibres length in each intervals (between 0^o and 180^o) and orientation of fibres in different parts of electrospun nanofibre mesh were measured to create a three dimensional model similarly as in the real sample by ABAQUS software (Fig. 3). The fibre was modeled as wire in software. A wire is depicted as a line in ABAQUS /CAE and used to idealize a solid in which it's diameter is considered smaller than its length. It is less expensive in comparison with other features like solid.

The interaction between adjacent fibres is modeled by Multi Point Constrain (MPC). A MPC tie constrains the motion of slave nodes of a region to the motion of a point. This constrain tie the fibre at the cross-section to each other. This is like a real fibre network, whereas spun fibres joined to each other as a result of residual solvent.

An Encastre boundary condition was applied in one side of structure, and the end elongation was applied in another side, similarly as in the tensile test. The Encastre boundary condition acts to restrain all degrees of freedom (Fig. 3).

2.3.2 Formulation of Model

The elastomeric PU has hyperelastic stress-strain properties[9]. Hyperelastic materials are described in terms of a "strain energy potential", which defines the strain energy stored in the material per unit of reference volume as a function of the strain at that point in the material. The hyperelastic material model is valid for materials that exhibit instantaneous elastic response up to large strains and requires that geometric nonlinearity be accounted for during the analysis step since it is intended for finite-strain applications[13]. In this study, the energy potential function of nanofibre yarns in the form of third order reduced polynominal function is used for the characterization of PU nanofibre as shown below:

$$U = C_1(I_1 - 3)^1 + C_2(I_1 - 3)^2 + C_3(I_1 - 3)^3 \quad \ldots (1)$$

where U is the energy potential function; C_I, the material constants; and I_1, the strain invariant. This form is used as yarn energy function (U_y), as given below:

$$U_y = 0.32(I_1 - 3)^1 - 1.04(I_1 - 3)^2 + 0.7(I_1 - 3)^3 \quad \ldots (2)$$

A static general method in which the effects of any nonlinearities presented in the model can be included, is used in the process of analysis. In this analysis, there is sequentiality so that the starting condition for each general step is the ending condition of the last general step, with the state of the model evolving whole history of general analysis steps as it responds to the history of loading.

A 2-node linear beam in space, hybrid formulation (B31H), is used as the mesh. Hybrid beam element type is provided in ABAQUS for use in the cases where it is numerically difficult to compute the axial and shear forces in the beam by the usual finite element of displacement method. This problem arises most commonly in geometrical nonlinear analysis when the beam undergoes large rotations and is very rigid in axial and transverse shear deformation. The problem in such cases is that slight differences in nodal positions can cause very large forces, which in turn cause large motions in other directions. To overcome this problem, hybrid elements use a more general formulation in which the axial and transverse shear forces in the elements are included as primary variables along with the nodal displacements and rotations, as primary variables[13].

2.3.3 Comparison of Model to Experimental Measurements

Three parameters are necessary to compare the model's predictions of the elastic modulus with those measured experimentally for PU meshes, namely fibre hyper elastic potential function, fibre diameter, and the network orientation. Fibre diameter and the network orientation were calculated from experimental measurements of fibre, and the fibre hyperelastic potential function (U_f) was determined by fitting the model to the experimental stress-strain response for meshes electrospun from 14.0 wt% solution. The energy potential function of fibre was obtained through comparison between experimental and modeling value.

3 Results and Discussion

The experimental results show that fibre diameters increase with increasing electrospinnig duration, polymer content and DMF content in THF/DMF mixture. The results also show that thickness and tensile properties of PU mat increase with increasing electrospinning duration. These results also show that with increasing THF content, tenacity and elongation of layers decrease.

The computational model was fit to the stress-strain curve for a PU mesh electrospun from a 14.0 wt% solution (Fig. 3). The resultant best fit for the fibre potential energy(U_f) was calculated using the following equations:

$$U_f = 0.37(I_1 - 3)^1 - 1.35(I_1 - 3)^2 + 2.7(I_1 - 3)^3 \quad \ldots (3)$$

Figure 4 shows the experimental and theoretical curves for comparison. Using the stress-strain data of electrospun PU meshes, the potential energy function

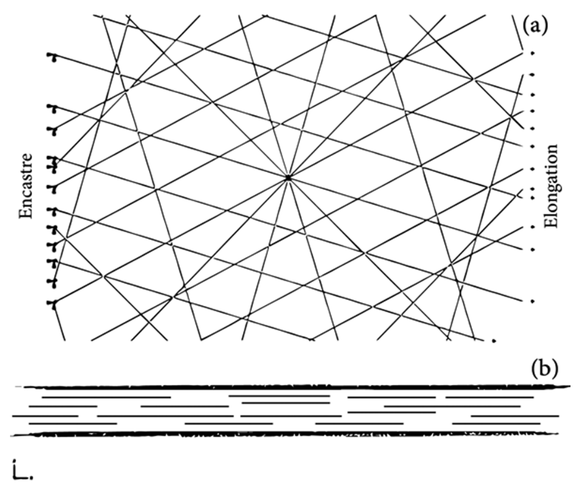

Fig.3—Representation of 3D fibre meshes (a) in x-y plane, and (b) in x-z plane

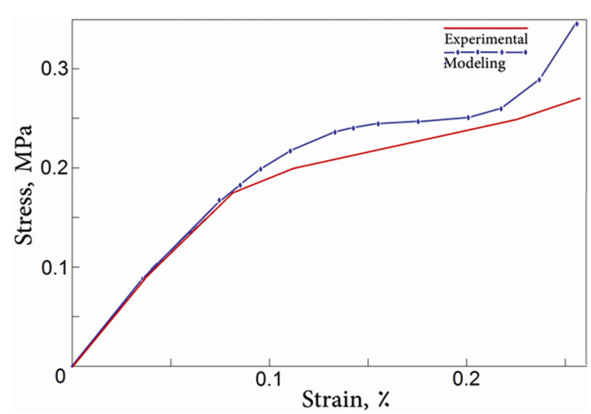

Fig. 4—Modeling (dot-line) and experimental (line) curves of PU mesh

of mesh (U_m) was obtained with least square method with the keep of following equation:

$$U_m = 0.28(I_1 - 3)^1 - 0.058(I_1 - 3)^2 + 0.011(I_1 - 3)^3 \quad \dots (4)$$

The obtained value of I_1 for fibre (Eq.3) is 0.37 and for fibre mesh is 0.28. This value determines the slope of the tensile curve in low strain and it can be assumed as young modulus. As the data show, this value for fibre is 48% higher than fibre network. This is because of volume fraction and network effects. Molnar *et al.*[14] reported that the difference between modulus of fibre and network of electrospun polyamide is 48%, and Stylianopoulos *et al.*[9] reported this difference about 26% for PU electrospun nanofibre and PU fibres network.

The tensile behavior of electrospun PU webs was measured as a function of fibre diameter, fibre orientation and network thickness, and then compared with proposed model for fibrous networks. The experimental results reveal that the tensile properties of fibrous meshes are sensitive to the microstructural architecture.

The obtained results from FEM model show that strength of fibrous network increases with increasing the fibre diameter, which is in accordance with the experimental results (Table 2). The Pearson correlation test shows a 90% correlation between modeling and experimental results that are in accordance with the results obtained by other researchers [10].

The tenacity in the machine direction of web is less than that in transverse directions of them. This difference comes from the more fibre that is oriented in the traverse direction of layer. Strength in different directions of electrospun nanofibre webs is due to the orientation distribution of the nanofibres in various directions[15]. It is thought that the accrued difference in this work is due to the traverse motion of the drum that causes the orientation of fibre bundle in cross direction. Table 3 shows the effect of fibre orientation (direction of loading) on tensile strength for experimental and modeling whereas there is an acceptable correlation among the results of these both methods (Table 3).

Statistical analysis shows significant difference between strength of fibre network for different levels of electrospinning duration (2, 4 and 6 hours). With increasing process duration, weight, thickness and strength of layers increase. Figure 5 shows the

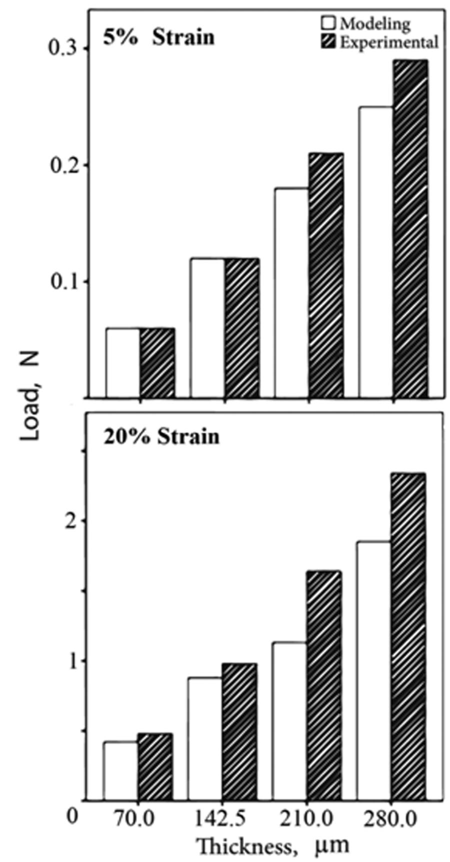

Fig. 5—Load versus thickness of fibre network for experimental and computed at 5% and 20% elongation

Table 2—Effect of fibre diameter on tensile strength at different stains

Fibre diameter nm	Load, N (5% strain)			Load, N (20% strain)		
	Modeling	Experimental	Error %	Modeling	Experimental	Error %
61	0.050	0.058	17	0.500	0.582	20
81	0.060	0.066	9	0.500	0.550	9
95	0.068	0.076	10	0.670	0.620	8

Table 3—Effect of fibre orientations on strength

Load directions	Load, N (5% strain)			Load, N (20% strain)		
	Modeling	Experimental	Error %	Modeling	Experimental	Error %
MD	0.053	0.056	23	0.358	0.565	25
CD	0.052	0.075	29	0.517	0.588	29

modeling results can accurately predict the effect of network thickness on tensile strength. The characteristics that distinguish the presented model from other models are (i) it incorporates fibre-fibre interaction; (ii) it accounts for three-dimensionality of PU fibre meshes, (iii) it can predict the mechanical behavior of the PU fibre network at large strain, and (iv) it considers PU fibre as hyperelastic material.

4 Conclusion

The proposed model in this study is based on the finite element method that has been developed for the tensile behavior of electrospun nanofibres. The experimental results indicate that there is a relation between the fibre diameter, fibre orientation and the tenacity of electrospun nanofibre mesh. Hence, the increase in fibre diameter and fibre orientation increases the tenacity of electrospun nanofibre mesh. The results obtained from modeling are in accordance with the experimental results. It is possible to predict the FN mechanical properties and optimization of FNs with strong contact bounds from this proposed model. Since networks with realistic geometries are applicable in this modeling, this analysis can ultimately serve as a tool for understanding the way in which mechanics of fibres and cross-links at the microscopic level determine the macroscopic behavior of the network.

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