Iranian Journal of Materials Forming 10 (3) (2023) 36-42

Online ISSN: 2383-0042

Iranian Journal of Materials Forming

Journal Homepage: http://ijmf.shirazu.ac.ir

Research Article

A Model to Estimate the Stress-Strain Behavior of Polyimide Electrospun Fabric

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ARTICLE INFO

Article history:

Received 09 July 2023
Reviewed 26 September 2023
Revised 03 February 2024
Accepted 04 February 2024

Keywords:

Modeling
Nonwoven
Polyimide
Mechanical properties

Please cite this article as:

S. Parsaei, S.M. Zebarjad, N. Anjabin, M.H. Moghim, A Model to Estimate the Stress-Strain Behavior of Polyimide Electrospun Fabric, *Iranian Journal of Materials Forming*, 10(3) (2023) 36-42.

ABSTRACT

Nonwoven structures consist of fibers with different orientations. Studying the mechanical behavior of nonwoven materials is very complicated and laborious in terms of the random nature of their constituent fibers. In this study, a suitable approach based on existing models is proposed to predict the stress-strain behavior of electrospun polyimide (PI) non-woven fabric as a function of the volume fraction, orientation distribution, and stress-strain behavior of its constituent fibers. 18 different discrete orientations from 0 to 180 degrees are considered to specify the fiber orientation distribution in the fabric. To avoid difficult and complex experiments on the fibers constituting the nonwoven fabric, the constants and characteristics of the stress-strain curve of a single fiber were determined by fitting the fabric stress-strain curve predicted by the model to the results of the fabric experimental tensile test. The comparison among the predicted stress-strain curves by the model and the experimental results for PI nonwoven fabric in two different loading directions of 0 and 45 degrees shows the validity of the method used in obtaining the stress-strain behavior of the fabric.

1. Introduction

Nonwoven fabrics are a type of advanced sheet materials or network structures that are made by connecting fibers or threads by mechanical, thermal, or chemical bonding methods [1, 2]. Woven fabrics are often used in clothing and general fabric materials as they are more cost-effective than non-woven fabrics and are easy to produce. Weaving multiple yarns together creates woven fabrics. In most cases, woven fabrics have higher strength values than non-woven fabrics. Non-woven fabrics have many superior properties to woven; including excellent thermal insulation and resistance to bacteria [1]. Nowadays using nonwoven fabric in some applications such as scaffolding, water purification membranes, and battery membranes is very common. Non-woven textiles are produced in different ways such as thermal bonding, needling, and melt blown process [3]. Electrospinning is one of the most important methods for producing non-woven [4-6].

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https://doi.org/10.22099/IJMF.2024.47803.1265

In order to improve the performance of these textiles, there is a need to find a correlation between the macroscopic behavior with their microstructure and the properties of electrospun fibers. Tensile properties play a key role in organizing and optimizing the mechanical properties of nonwovens [7]. Considering the challenges, we face in the experimental measuring of the mechanical properties of nanofiber mats, it is desirable to develop a model for the tensile properties of nanofiber mats. On the other hand, the tensile properties of the mats can be used as a criterion for the mechanical properties of a single nanofiber [8]. Because of the complicated behavior and random nature of the constituent fibers, modeling the mechanical behavior of electrospun materials is a challenging task. Since the significant deformation, rotation, bending, breaking, and slippage of fiber are common during mechanical loading of nonwovens [9].

To the best of our knowledge, the modeling of mechanical behavior of nonwoven fabric can be categorized into four groups. The first group predicted the elastic behavior of paper based on the mechanical characteristics and distribution of its constituent fibers. The fibers were considered as continuous threads of filaments, and their deformation involved only the axial stretching. Other deformation modes such as bending, breaking, and slipping of the fibers did not enter the model. For example, Cox [10] proposed a model to analyze the mechanical behavior of nonwoven fabrics with a structure similar to electrospun materials. The second group used the well-known orthotropic theory to predict the mechanical behavior of nonwoven fabrics. In this model, the nonwoven structure is supposed to be a layered structure and each layer is considered as a lamina. For given values of the elasticity modulus, shear modulus and Poisson's ratio in the two mutually perpendicular principal directions of the material, the orthotropic theory can be used to predict the stress-strain behavior of the material when subjected to loading in any direction. While this theory is only valid for the small elastic strains, it can partially predict the nonlinear properties of oriented materials [11]. For example, Backer and Petterson [11] used this model to predict the stress-strain curve of non-woven fabric and observed a good agreement between the model prediction and the experimental curve. The third group concentrated on fiber network theory which included structural characteristics, and suitable non-woven structures. In this model, it is assumed that the non-woven fabric is composed of many small components, and the fibers are bonded at the border of each component. Each component experiences a strain equal to the strain of the fabric. Additionally, an average strain is created in each fiber according to the connection points. The disadvantage of this method is that the number of connection points is not specified. These connection points are important because they transfer load, and their number in a given volume has been presumed by researchers. In this theory, the fibers between the connection points were assumed to be straight without any curvature. This model was later extended by adding the effect of fiber curvature and the curvature distribution of the fibers by Hearl and Stevenson [12, 13]. The fourth group tried to use finite element models to predict the mechanical behavior of nonwoven fabric.

The simulation of the mechanical behavior of nonwovens is divided into continuous, and discontinuous models via the finite element. In discontinuous models, the fibers are precisely modeled in a specific pattern, and their irregularity is applied in the model [12, 13]. Silberstein et al. presented a discontinuous model to predict the macroscopic behavior of nonwovens [14]. Using a homogenization approach and a multi-layered triangular mesh, their model predicted the behavior of the material under cyclic and uniform loads. In continuous fiber models, non-woven network is considered as a continuous medium, and mechanical properties are simulated by assigning properties to the elements [13]. The shape and size of the connection points are precisely entered in this model, and the randomness of the fiber orientation (visible in the orientation distribution function) is calculated using the software. This model was used to predict the stress-strain behavior of nonwovens with high density [9, 13]. The main limitation of this method is the lack of information about the microstructure mechanism, which cannot be studied with this method. However, the advantage of this method is that it is computationally efficient for predicting macroscopic behavior [13].

Bais-Singh and Goswami considered a continuous composite structure based on the laminate composite model and introduced the effect of non-uniform fiber orientation [15]. In this method, it was assumed that the non-woven fabric is continuous and consists of different layers. All the fibers of one layer have the same direction, and there is an angle between the direction of the fibers of one layer and the other layer, which generally indicates the anisotropy of the desired texture [13]. In the present study, the Bais-Singh and Goswami's model has been used to predict the tensile behavior of PI electrospinning fabric.

In this model, constants of the stress-strain curve of a single fiber of the fabric are needed but obtaining the stress-strain curve of a fiber is a challenge due to the laboratory limitations for tensile testing of a single nano sized fiber. To overcome the laboratory challenge of measuring the stress-strain curve of a single fiber, the model constants that characterize the stress-strain behavior of a single fiber were estimated by fitting the model predictions on the experimentally measured stress-strain curve along one direction of PI electrospun fabric. Then the model was validated by predicting the tensile behavior of the fabric in other directions.

2. Theoretical framework

The study of tensile behavior is one of the most common methods to characterize the mechanical behavior of nonwovens. Therefore, despite the complexity of the behavior of nonwovens, it is very useful to use the model to predict their tensile behavior.

2.1. The Bais-Singh and Goswami's model

Bais-Singh and Goswami used Equation 1 to calculate the stress-strain relationship of the nonwoven fabric in a given tensile direction [2, 6]. All fibers are assumed to be straight. Where T(α) is electrospun fabric tensile stress in the tensile direction (α). σ^f and ε_f are fiber stress and strain, respectively, which is related to each other through Equation 2. V_f is volume fraction of fibers, β_i is the angle of the fiber with respect to loading direction, and X (β_i) is a fraction of fiber repetition in the β direction. It should be noted that e is the fabric strain, which is related to the fiber strain in Equation 3.

$$T(\alpha) = \sum_{i=1}^{n} \sigma^f cos^2 \beta_i V_f X(\beta_i) \tag{1}$$

$$\sigma^f = f(\varepsilon_f) \tag{2}$$

$$\varepsilon_f = g(e) \tag{3}$$

In this model, the nonwoven fabric is assumed to be a set of fibers in which each fraction of fibers (X) is positioned in a certain direction (β_i) relative to the loading direction (α). To simplify the understanding of the model based on Fig. 1, the fabric can be considered as a set of n layers where the fibers in the i^{th} layer are located in a certain direction β_i and the tensile load is applied to the fabric at an angle α to the x-axis. A prerequisite for using above theory is to know the fiber stress-strain relationship and the orientation distribution of the fibers.

2.2. Relation between fiber strain and non-woven fabric strain

To make the calculations easier, it is assumed that when electrospun fabric are tensioned; strain will appear only in the direction of applied stress and the strain along the transverse direction can be neglected. Fig. 2 shows how the length of a fiber changes after stretching nonwoven fabric, assuming no transverse strain.

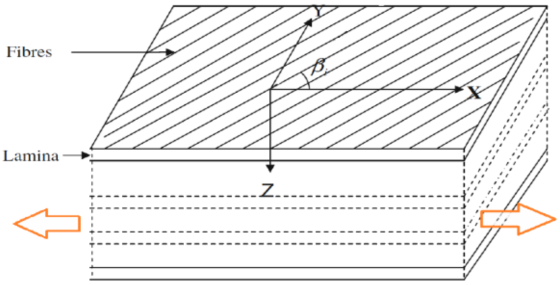
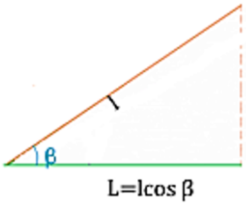


Fig. 1. Non-woven fabric in the form of a multilayer laminate [6].



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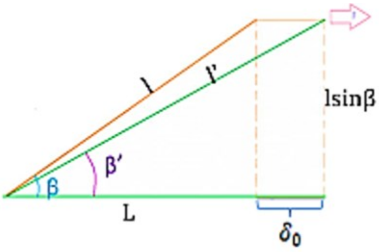

After

Fig. 2. Fiber length before and after stretching (assuming no transverse strain).

Based on Fig. 2, the initial length of the fiber before stretching is l and, l', the length of the fiber after stretching is obtained according to Pythagorean law .As shown in the Fig. 2; $l = L/\cos\beta$. Also, fabric strain (e) is δ_0/l . Subsequently, the fiber strain (ε_f) in terms of fabric strain (e) is obtained according to the following geometric equations.

$$l' = \sqrt{(l\sin\beta)^2 + (l\cos\beta + \delta_0)^2}$$

$$\varepsilon_f = \frac{l' - l}{l}$$

$$\varepsilon_f = \frac{\left(\sqrt{(l\sin\beta)^2 + (l\cos\beta + \delta_0)^2} - l\right)}{l}$$

$$\varepsilon_f = \sqrt{\sin^2(\beta) + (\cos\beta + \frac{\delta_0}{l})^2} - 1$$

$$\varepsilon_f = \sqrt{\sin^2(\beta) + \cos^2(\beta) + \left(\frac{\delta_0}{l}\right)^2 + 2(\frac{\delta_0}{l})\cos\beta} - 1$$

$$as \qquad \sin^2(\beta) + \cos^2(\beta) = 1$$

$$\varepsilon_f = \sqrt{1 + \left(\frac{\delta_0}{l}\right)^2 + 2\left(\frac{\delta_0}{l}\right)\cos\beta} - 1$$

$$as \qquad l = L/\cos\beta$$

$$\varepsilon_f = \sqrt{1 + \left(\frac{\delta_0}{L}\right)^2 \cos^2(\beta) + 2\left(\frac{\delta_0}{L}\right)\cos^2(\beta)} - 1$$

$$as \qquad e = \delta_0/l$$

$$\varepsilon_f = \sqrt{1 + e^2\cos^2(\beta) + 2e\cos^2(\beta)} - 1$$

$$\varepsilon_f = \sqrt{1 + \cos^2(\beta)(e^2 + 2e)} - 1 \qquad (4)$$

2.3. Stress-strain behavior of a single fiber

Based on the experimental data reported in the published literature for fiber stress-strain behavior [16-18], in this study the stress-strain relationship of a single fiber was considered to be bilinear, as follows.

$$\boldsymbol{\sigma}^f = \begin{cases} E_f\varepsilon_f & \sigma < \sigma_y^f \\ \sigma_y^f + H\left(\varepsilon_f - \varepsilon_f^y\right) & \sigma > \sigma_y^f \end{cases} \qquad (5)$$

Where ε_f^y and σ_y^f are fiber yield strain and fiber yield stress, respectively. E_f and H are constants. Since it is difficult to prepare a single PI fiber and perform tensile testing on it because it requires advanced equipment, the model constants appearing in Equation 5 were estimated through curve fitting on the experimental fabric tensile test data. By substituting Equations 4 and 5 into Equation 1, a complete model is obtained to describe the stress-strain relationship of nonwoven fabric during tensile loading.

3. Experimental Procedure

3.1. Materials

Polyimide powder (Matrimid5218) was purchased from Huntsman Company (USA). Solvents; dimethyl acetamide (DMAC) (99% purity) and ethanol (96% purity) were supplied by Samchun Company (South Korea) and Dr. Mojalalei Company (Iran) respectively. Solvents were used without any more purification.

3.2. Non-woven mat preparation

The polyimide powder was dissolved in DMAC solvent at a fraction of 16% by weight and stirred on a magnetic stirrer for 1 h to obtain a uniform solution. Electrospinning of PI solution was performed by an electrospinning machine (NanoAzmaCo. Iran). 5 ml of the solution was electrospun with a feed rate of 1.4 ml/h at a high voltage (17 kV) in a distance of 15 cm from the collector to the needle. The fibers were accumulated on a 100 rpm, rotating collector, covered by aluminum foil. All steps were done at ambient temperature (25 °C) and humidity of 30%.

3.3. Fiber orientation distribution

A scanning electron microscope (Cambridge-S360 model, 20 kV accelerating voltage) was used to obtain an image of the surface of the mat, in order to determine the fiber repetition fraction in the electrospun sample. Then, the number of fibers in each direction was counted using Image J software. It should be noted that the mat was coated with gold before SEM.

3.4. Volumetric fraction of fibers

The volume fraction of fibers is calculated using volume porosity via the following formula:

$$V_f = 1 - P \tag{6}$$

Where V_f volumetric fraction of fibers and P is volumetric porosity.

To measure the volumetric porosity of the produced mat, it was immersed in ethanol solvent for 2 hours after measuring the mass. The mass of mat after immersion was then measured by taking excess ethanol with filter paper. Finally, volumetric porosity was calculated using the following formula.

$$P = \frac{\left((M_{wet}-M_{dry})/\rho_{solvent}\right)}{\left(\frac{M_{wet}-M_{dry}}{\rho_{solvent}}\right)+\left(\frac{M_{dry}}{\rho_{polymer}}\right)} \tag{7}$$

Where M_{wet} and M_{dry} represent the mass of the mat before and after immersing for 2 hours in ethanol. $\rho_{solvent}(0.8\ \frac{gr}{cm^3})$ and $\rho_{polymer}(1.24\ \frac{gr}{cm^3})$ are Ethanol and Polyimide densities, respectively.

The tensile test was performed by a universal testing machine (Ghotech Co. Taiwan), and the samples required for tensile test were punched in strips with the width of 10 and a 20 mm gauge length. Crosshead speed of the tensile device was 20 mm/min. The thickness of produced math was 240 ± 35 microns.

4. Results and Discussion

To obtain fiber fraction (X (β$_i$)), three different SEM images were prepared from the electrospun mat. Then, using Image J software, the number of fibers in each direction was determined. A total of 18 different discrete orientations with an interval of 10 degrees from 0 to 180 degrees were considered. Fig. 3 shows an electrospun PI fabric SEM image and the fiber orientation distribution.

The volume fraction of fibers (V_f) has been calculated using porosity using equations 6 and 7 and the mentioned method in section 3. The volumetric porosity of the used sample is 0.973, therefore V_f =0.0262.

Equation 5 shows the general form of a bilinear equation for fiber stress-strain relationship. As explained before the model constants (E_f, H, and ε_f^y) were estimated by fitting the computed stress-strain curve of fabric with the experimental tensile test results performed at the x- direction (Fig. 1). The estimated values for constants of Equation 5 are shown in Table 1.

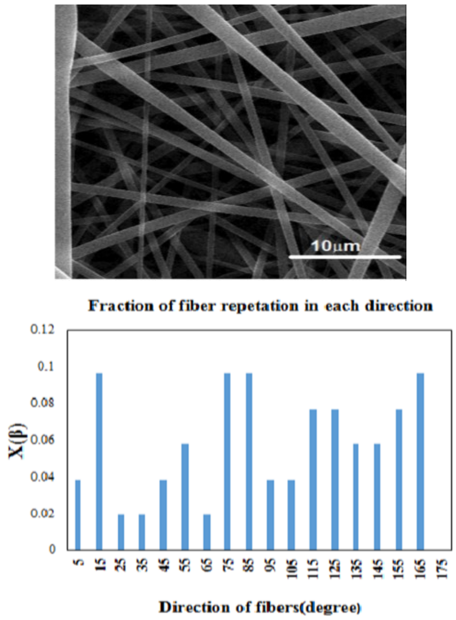

Fig. 3. SEM image of PI electrospun fabric and the distribution of fiber orientation.

Table 1. Estimated values of constants in Equation 5

Ef	H	Ɛfy
4.657 GPa	0.836 GPa	0.017

The predicted stress-strain curve of fabric loaded in x-direction are shown in Fig. 4, along with experimental data.

Using Equations 1, 4, and 5 and the obtained values in Table 1, the stress-strain curve of the electrospun fabric can be predicted in other directions.

Fig. 5 shows the predicted stress-strain curve for the fabric during the tensile test along the 45-degree relative to x-axis, with the corresponding experimental strain-stress data. The comparison of the predicted stress-strain curve and the experimental results confirms the validity and applicability of the proposed model to predict nonwoven fabric tensile behavior at different loading directions.

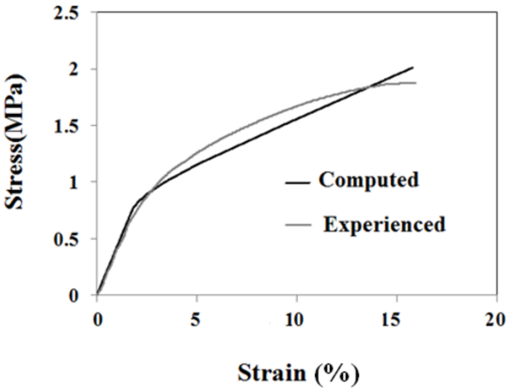

Fig. 4. Comparison between the predicted and experimentally measured stress-strain curves of nonwoven fabric loaded in x-direction.

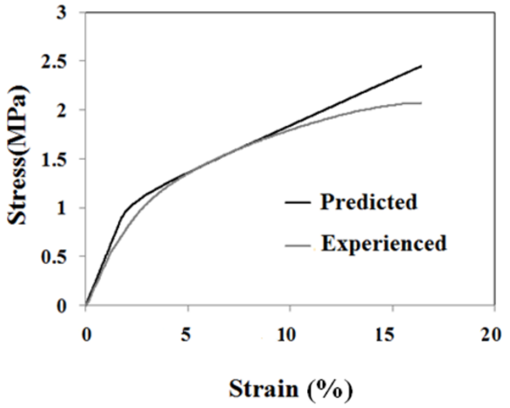

Fig. 5. Predicted and experimental stress- strain curves of non-woven fabric loaded at the 45-degree relative to x-axis.

5. Conclusion

The stress-strain behavior of polyimide electrospun nonwoven fabric was predicted during uniaxial loading as a function of fiber volume fraction, fiber orientation distribution, and loading direction.

- In the presented model, the stress-strain response of a single fiber was considered as a bilinear function, and the corresponding constants were obtained by fitting the model predictions on the experimental stress-strain curve of the fabric.
- The model predictions have a good agreement with the fabric stress-strain curve obtained in the laboratory for different loading directions.

Acknowledments

The authors are deeply grateful to all those who played a role in the success of this research.

Conflict of Interests

The authors declare no conflict of interest in this research.

Funding

This research has not received any funding.

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