CORRELATION ANALYSIS OF ELECTROSPINNING PROCESS PARAMETERS

Ph.D Iuliana **G. LUPU**[1], Ph.D **Oana CRAMARIUC**[2,3], Ph.D **Liliana HRISTIAN**[1], Ph.D**. Nicanor CIMPOEȘU**[1] , Ph.D **Marian C. GROSU**[4]

[1]"Gheorghe Asachi" Technical University from Iasi, Romania,[2] IT Center for Science and Technology, Bucharest, Romania, [3]Tampere University,Tampere, Finland, [4]The National Research and Development Institute for Textile-Leather, Bucharest, Romania

REZUMAT. În această lucrare au fost investigați doi parametri importanți ai procesului de electrofilare și anume debitul și distanța dintre vârful acului și colector, folosind o soluție polimerică PEO/etanol și apă. Analiza SEM a relevat faptul că fibrele electrofilate prezintă o morfologie uniformă cu o distibuție îngustă a diametrului fibrelor. Influența distanței vârf-colector respectiv a debitului asupra diametrului fibrelor electrofilate a fost investigată folosind analiza statistică. Astfel, analiza a indicat o dependență liniară între parametrii procesului și diametrul fibrelor. Acest lucru înseamnă că, odată cu creșterea distanței vârf-colector diametrul fibrei scade în timp ce creșterea debitului conduce la creșterea diametrului fibrelor electrofilate.

Cuvinte cheie: electrofilare, oxid de polietilenă, analiză statistică.

ABSTRACT. In this paper, we have studied two important parameters of the electrospinning process, including needle-tip-to-collector distance, and flow rate by using using PEO/ethanol and water polymer solutions. Scanning electron microscopy (SEM) analysis showed that the electrospun fibers have relatively uniform morphologies with a narrow fiber diameter distribution. The effect of tip-collector distance and flow rate on the fiber diameter was investigated by using statistical analysis. This revealed a liniar dependency between process parameters and fiber diameter. This means that with increasing distance tip-collector the fiber diameter is decreasing while with increasing of flow rate the fiber diameter is increasing.

Keywords: electrospinning, polyethylene oxide, statistical analysis

1. INTRODUCTION

Among various technologies, electrospinning which is a versatile technique known for decades [1-3], has lately attracted significant attention as a commercially viable route to fabricate micro or nano-fibers for a large number of industrial applications, including filtration media, electronics, biomaterials, and clinical medicine, etc.[4-6].

This method uses an electrostatic potential between two electrodes. The first electrode is a spinneret from which a liquid (polymer solution or melt) is drawn by the electrostatic field toward the second electrode, the collector [7, 8].

Polyethylene oxide (PEO) is a bio-friendly synthetic polymer, non-toxic and non-mutagenic, that has been extensively used for the production of electrospun fibers. It is usually dissolved in water or ethanol but also in organic solvents.

The electrospinning process is affected by a wide range of parameters, *i.e.* polymer concentration, applied voltage, flow rate, temperature, distance between tip and collector, electric field intensity, humidity, etc.

In this article, we report the synthesis and characterization of inexpensive fiber structures of micrometer fiber size that show potential for many applications. Scanning electron microscopy (SEM) was used to reveal the microstructure of the electrospun fibers.

To achieve a better understanding of the relationship between electrospinning parameters (distance between tip and collector and flow rate) and the morphology of the electrospun fibers, in particular the fiber diameter, this paper is using statistical analysis (linear regression).

2. EXPERIMENTAL

2.1. MATERIALS

Polyethylene oxide PEO solutions obtained from Aldrich were prepared at 5.5 wt% in deionized water and ethanol (> 99%, PanReac & AppliChem IWT Reagents) used as solvents without any further purification. Sodium chloride was used as additive to improve the electric conductivity of the polymer solution.

2.2. SOLUTION PREPARATION

PEO pellets (1.4 g) were added to a mixture of deionized water and ethanol in a weight ratio of approximately 4:1. An amount of 0.8 wt% of NaCl was added to the mixture to improve the conductive properties of the polymer. The solution was stirred for 24 hours using a magnetic stirring rod. A constant temperature of 26^0 C has been maintained throughout the study.

2.3. ELECTROSPINNING PROCESSION

The operation conditions of the electrospinning device for the preparation of the samples were as follows: the solution flow rate was 0.3-3mL/h1, applied voltage was adjusted to be 20 kV, and the distance between the syringe needle tip and collector was selected to be 50 – 70 mm.

The syringe with a needle inner diameter of 0.4 mm used in the experiments was driven by a syringe pump (Model: KDS 100, KD Scientific) which allowed control of the flow rate and volume of polymer ejected during each experiment. The syringe was fixed horizontally on the syringe pump and the solution was electrospun on a rotating drum by using a standard high voltage power supply (Matsusada HER 20R3).

Several tests with different process parameters were performed and those presented in Table 2.1 were selected for further investigation due to improved fiber properties, in special absence of beads.

Table 2.1. **Process parameters of selected samples**

Sample	Distance tip-collector (mm)	Flow rate (mL/h)	Voltage (KV)	Tip speed (mm/s)	Humidity (%)	Temperature (^{0}C)
S1	45	1	20	50	35	26
S2	60	1	20	50	35	26
S3	70	1	20	50	35	26
S4	45	0.3	20	50	35	26
S5	45	3.0	20	50	35	26

2.3. CHARACTERIZATION

The comercial aluminium foil used for collecting the fibers was kept to the established distance so that the solvent could evaporate and the fibers could get enought flight time to dry. These aluminium foils were used to collect the samples for visualization under the scanning electron microscope. The sample was placed in a SEM VEGA II LSH TESCAN and imaged with different magnification for dimensional characterization. For each image, at least 50 different points were randomly chosen to measure the average diameter of the fibers.

3. RESULTS AND DISCUSSION

The SEM images at 2000x magnification of the studied samples are given Fig. 3.1.

It is hard to draw a comparative conclusion between these five samples just by visual inspection, so an investigation of the fiber diameter distribution has been done.

For this purpose, a series of minimum 30 measurements were performed on the 2000x magnification SEM images of the samples in order to determine the diameter distribution. The histograms were obtained from the diameters and are shown in Figures 3.2 to 3.6. It can be seen that the fibers showed a random distribution forming three-dimensional fibrous structures. The PEO electrospun fibers have relatively uniform morphologies with a narrow fiber diameter distribution, indicating a viable and reproducible process.

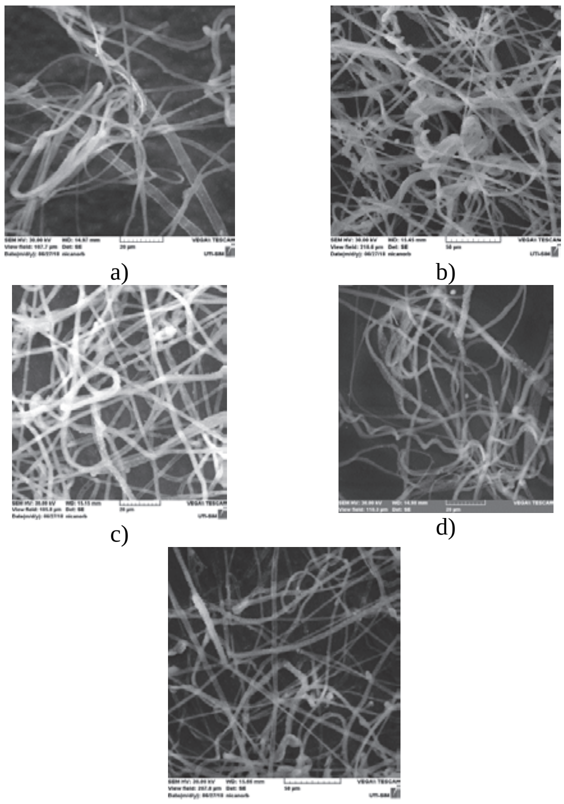

Fig. 3.1. SEM images at 2000x for: S1(a) , S2 (b), S3 (c), S4 (d) and S5 (e)

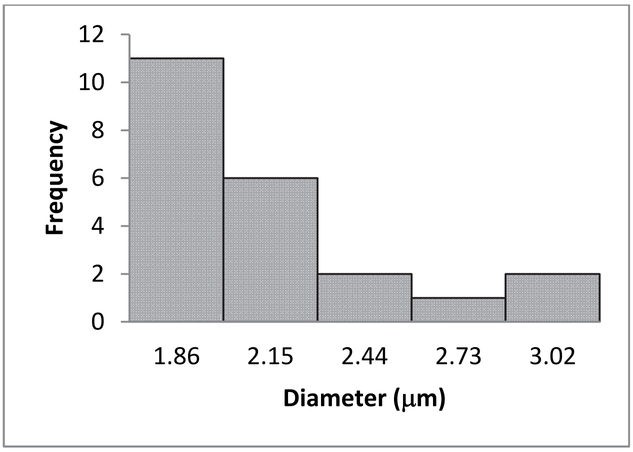

Fig.3.2. Diameter distribution of sample S1

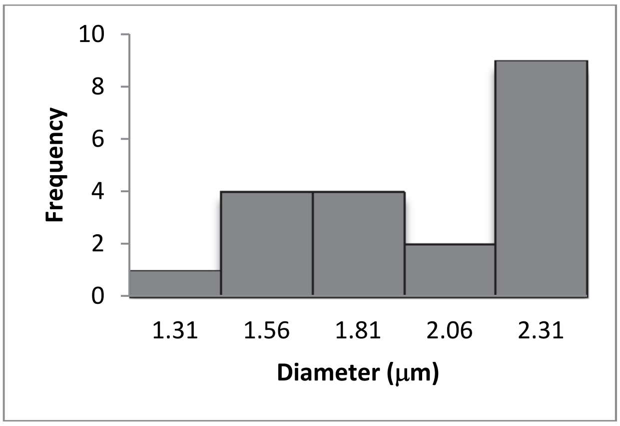

Fig.3.3. Diameter distribution of sample S2

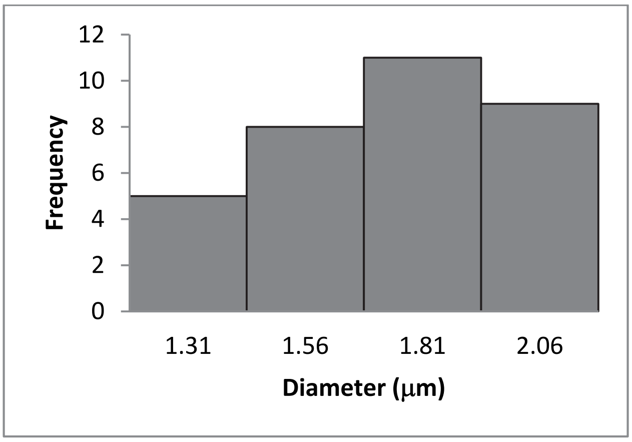

Fig.3.4. Diameter distribution of sample S3

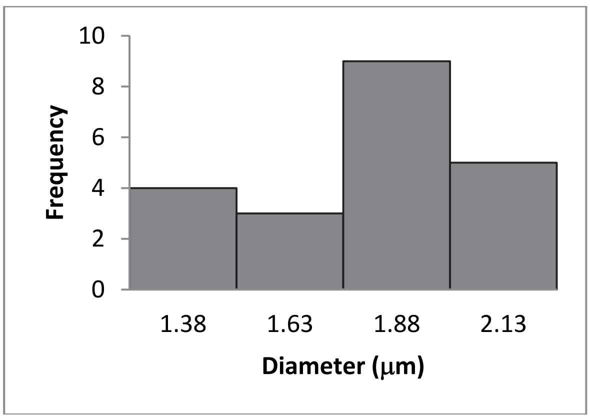

Fig.3.5. Diameter distribution of sample S4

We used statistical analysis (simple linear regression) in order to evaluate the dependency between flow rate and distance tip-collector, on one hand, and the fiber diameter on the other hand, by calculating the correlation coefficient between these variables.

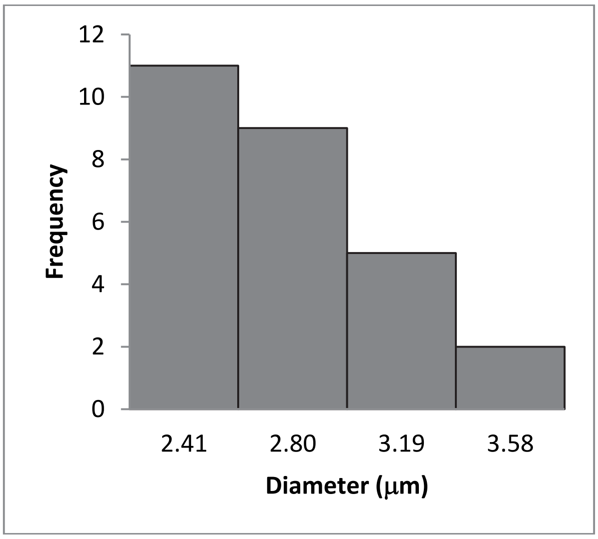

Fig.3.6. Diameter distribution of sample S5

The results are presented in Figures 3.7 and 3.8. A linear dependency exists between distance tip-collector, flow rate and the fiber diameter.

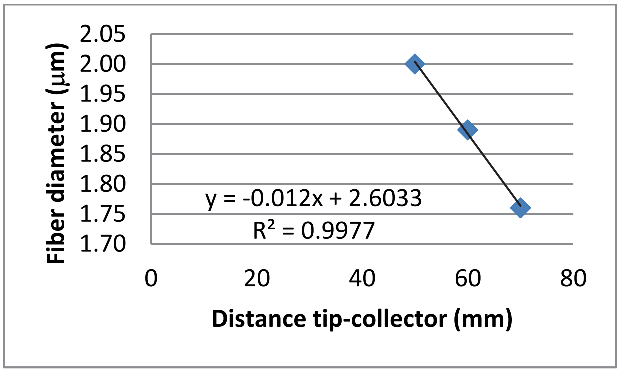

Fig.3.7. Dependency of distance tip-collector on fiber diameter

In both cases, the value of the correlation coefficient illustrates a very good and significant correlation (*i.e.* the value of R2 is near 1).

As can be seen from Fig.3.7 the larger the distance between tip and collector, the lower is the fiber diameter which is in line with previous studies [9,10]. With an increase in the distance of the fibers movement towards the collector, the electric field strength has the opportunity to stretch more the fibers and thus produces thinner fibers (2.0 μm for sample S1 to 1.76 μm for S3) .

An increase in flow rate from 0.3 mL/h for S4, to 1mL/h for S1 and then to 3mL/h for S5 (see Table 2.1) increased the amount of material flowing through the tip, which in turn resulted in increased fiber diameters (1.65 μm for S4 to 2.80 μm for S5).

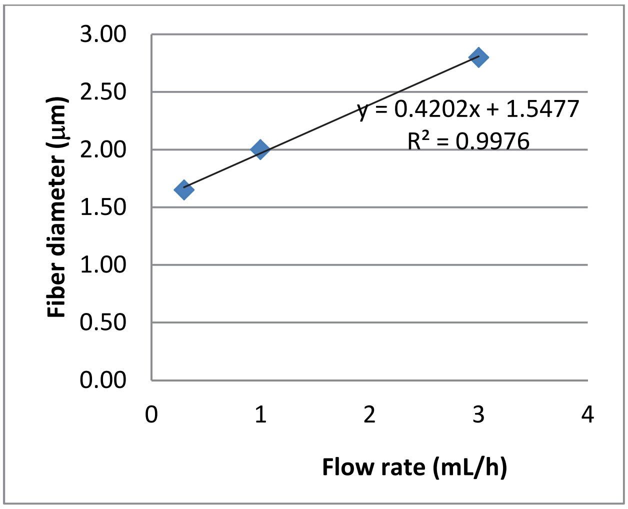

Fig.3.8. Dependency of flow rate on fiber diameter

4. CONCLUSIONS

Electrospinning is a versatile and low cost method for the production of micro and nano-fibers. In this study, the effect of different parameters was investigated as follow: distance tip-collector and flow rate on the diameter of electrospun PEO fibers was investigated. Statistical analysis revealed that the increase of distance tip-collector decreased the average micro-fibers'diameter. Inversely, the diameter of microfiber increased when flow rate increased.

ACKNOWLEDGEMENTS

This work was supported by the national grant no. 109PED/2017, MAGNANO, financed by Unitatea Executiva pentru Finantarea Invatamantului Superior, a Cercetarii, Dezvoltarii si Inovarii, Romania.

REFERENCES

[1] Doshi,J., Reneker, D. H., *Electrospinning process and applications of electrospun fibers*, J. Electrostat., vol. 35, no. 2/3,151-160, 1995

[2] Reneker, D. H., Chun, I., *Nanometre diameter fibres of polymer produced by electrospinning*, Nanotechnology, vol. 7, no. 3, 216-223, 1996

[3] Reneker, D. H., Fong, H., *Polymeric Nanofibers*, Washington, DC, USA:Amer. Chem. Soc., vol. 918, 2006

[4] Xu, J. et al., *Roll-to-roll transfer of electrospun nanofiber film for high-efficiency transparent air filter*, Nano Lett., vol. 16, no. 2, 1270-1275, 2016

[5] Ahmed, F. E., Lalia, B. S., Hashaikeh, R., *A review on electrospinning for membrane fabrication: Challenges and applications*, Desalination, vol. 356, 15-30, 2015

[6] Zheng, J. et al., *4-Dibenzocyclooctynol (DIBO) as an initiator for poly (ε-caprolactone): Copper-free clickable polymer and nanofiber-based scaffolds*, Polym. Chem., vol. 4, 2215-2218, 2013

[7] Demir, M.M., Yilgor, I., Yilgor, E., and Erman, B., *Electrospinning of polyurethane fibers*, Polymer, vol.43, 3303-3309, 2002

[8] Ibrahim, S., Sayed, H.M.,. EL-Rafei, A.M., El Amir, A., Ismail, M. and Allam, N.K., *Improved genistein loading and release on electrospun chitosan nanofiber blends*, J. Mol. Liq., vol. 223, 1056-1061, 2016

[9] Jabur, A.R., Abbas, L.K, Muhi Aldain, S.M., *Effect of ambient temperature and needle to collector distance on PVA nanofibers diameter obtained from electrospinning technique*, Engineering and Technology Journal, vol. 35, Part A, no.4, 2017

[10] El Gohary, M. I. et. al., *Controlling the Features of Electrospun Nanofibers*, Egypt. J. Biophys. Biomed. Engng, vol.18,13-21, 2018

About the authors

Ph.D **Iuliana Gabriela LUPU**
Technical University of Iasi, Romania

Graduated the Technical University from Iasi, Textile-Leather Faculty in 1992. In present, employed at Technical University from Iasi where currently holds a position of associate professor. Has a Ph D degree since 2002 in Industrial Engineering. Areas of competences: nonwovens, technical textiles, electrospinning

Ph.D **Oana CRAMARIUC**
IT Center for Science and Technology, Bucharest, Romania, Tampere University,Tampere, Finland

Holds a M.Sc. degree in Material Sciences and organic chemistry and a PhD degree since 2006 in physics from Tampere University of Technology, Finland where holds the title of adjunct professor. Areas of competences: modeling tools for the investigation of complex phenomena and compounds, simulation, electrospinning

Ph. D **Liliana HRISTIAN**
Technical University of Iasi, Romania

Graduated of the Technical University "Gheorghe Asachi" in Iași, Faculty of Textiles - Leather - 1997, Ph D degree since 2008, lecturer at the Faculty of Textile Leather and Industrial Management - 2012. Areas of competence: testing and quality control of textile products, methods processing of statistical data.