WIRE SPINNER FOR COAXIAL ELECTROSPINNING

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Abstract

The paper focuses on the problematic of coaxial electrospinning with a wire spinning electrode. Despite the wide usage of wire spinner in the industrial sector, only basic research has been made in the field of possible coaxial technology application. In the paper, the new concept of the wire spinner, reflecting the polymeric two-layer formation, is introduced. The design is supported by electrostatic analysis of the problem using FEM method. The manufactured testing device was experimentally verified and the results were evaluated. The electrospinning process stability and the final product were analysed and the potential of the application was discussed.

Keywords: Wire electrode, core/shell nanofibers, coaxial electrospinning, electrostatic analysis

1. INTRODUCTION

Out of the many ways of producing nanofibers [1], an electrospinning is the most widely used technology. It is mainly because of its wide range of materials spinnable by the electrospinning process. Basically, there are two types of electrospinning technologies: the DC electrospinning, using bi-polar set-up [2], and recently published AC electrospinning, where only one electrode run by alternate current is needed [3]. For each of these methods a variety of spinnerets can be used, based on the required type of nanofibers produced or on the productivity of the device [1]. Out of these different spinnerets designs, few of them have been used in the industrial sector, while others remain in the laboratory scale usage. First type of spinneret with the potential of industrial usage, invented by Jirsak et al, is based on the cylinder rotating in the polymeric solution [2]. Another one is based on the usage of wire spinner [4].

The advanced application of electrospinning technology is related to the production of core/shell nanofibers. These fibres consist of core and shell polymer of different properties. These nanofibers have a high potential of use in a tissue engineering to replace damaged tissue or as materials for a drug delivery system [5]. Production of such fibres requires formation of double polymer droplet/layer. During the electrospinning, where the Taylor cones and polymeric jets are formed, the outer polymer layer creates the shell of the fibre, while the inner one is drawn into the fibre as its core.

The first equipment for production of core/shell nanofibers using needless technology with potential use in the industrial sector was developed at Technical University in Liberec and patented in 2009 [6]. This equipment called “weir spinner” uses the principle of the overflow thin polymer two-layer over the edge of spinning electrode. Forward et. al. introduced the coaxial wire electrospinning from a free surface of polymer solution in 2013 [7]. The metal wire electrodes are wading through the polymer two-layer. The thin polymer bi-component film is created on the wire and formed to core/shell nanofibers by applied high voltage. The advantage of the wire spinner is based on its simple design and optimal distribution of electric intensity over the wire length. Thanks to relatively low diameter of used wires, the electrical intensity concentrated on the wire surface reaches high values under relatively low voltage applied. There were several studies of using wires of different properties and shapes [8]. Introducing of the coaxial fibre production to the wire spinner brings several challenges related to the polymer solutions dosage to the wire and with a homogeneous distribution of polymer two-layer throughout the electrospinning process. However, further study and analysis verified by carried out experiments can contribute to the successful implementation of the coaxial production to the wire spinner technology. This will broaden the usage of the wire spinner technology in the nanofiber production.

2. TESTING DEVICE

In order to study and test the electrospinning process, a testing device with the wire electrode was designed and manufactured. The device was designed for a low-scale laboratory production of nanofibers and fully reflects the requirements for variability of main parts of the device for different experiments. The scheme of the designed wire testing spinner is shown in the **Fig. 1**. This concept is based on integrated winding system for winding a wire electrode *1*. This electrode is connected to the positively charged power source. At the certain distance from the wire electrode, there is a collector *2*, connected to the negatively charged power source.

The key part of the device is a system for applying of the polymer solution on the wire. This is performed by the static polymer dosing cartridge *3*. This subsystem provides the required amount of dosed polymer solution to be applied on the wire that is drawn through the cartridge. In order to optically control the process of polymer application, the cartridge is made of transparent material. The cartridge with two inputs provides the application of two polymer solution, creating the double polymer layer. The double-polymer layer, drawn out from the cartridge is then electro-spun in the active zone. This zone is placed in between the pulleys and under the collector, and is characterised by high electric intensity on the polymer surface that enables the start of the electrospinning process. The position of the wire in the active zone is guided by the dosing cartridge on one side and a guiding part *6* on the other side. The polymer solution for the core (P_c) and the shell (P_s) is transported to cartridge inputs in hoses from external peristaltic pumps. By control of the amount of dosed polymer, the thickness of the polymer layer on the wire can be regulated.

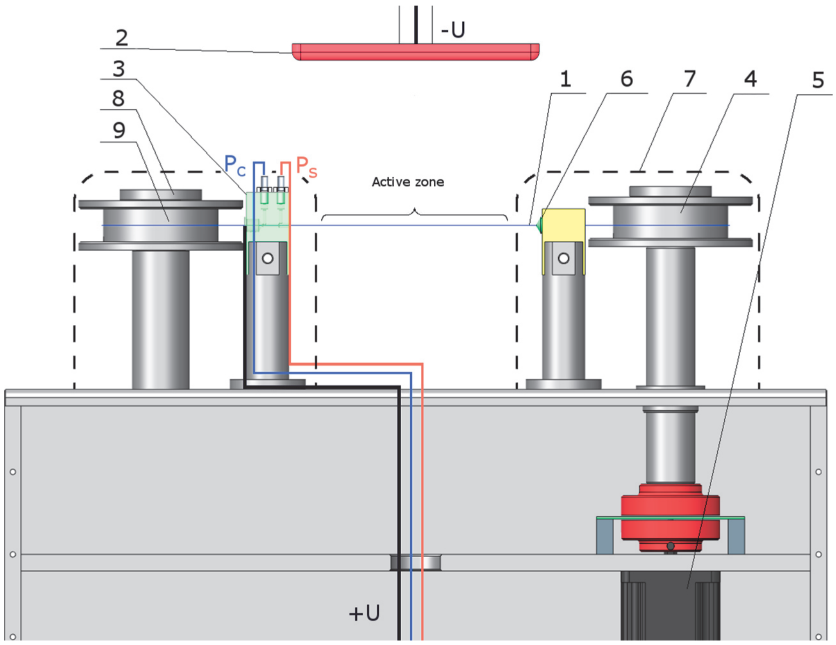

Fig. 1 The scheme of the wire spinner, 1: Wire electrode (+U), 2: collector (-U), 3: Polymer solution (s) dosing cartridge, 4: Driven pulley, 5: Step motor, 6: Wire guide, 7: Covers, 8: Friction brake, 9: Free pulley

The winding system of the device enables winding the wire of variable diameter and material from a freely rotated pulley *9* to the driven pulley *4*. The winding speed can be regulated within certain range by means of step motor controlling system *5*. The required wire tension during the winding process is provided by means of friction brake *8*, placed on the top of the freely rotated pulley *9*. The designed system enables winding of stored bobbin of wire with wire diameter up to 0.5 mm. Furthermore, by replacing of guide parts with bigger inner diameter, this can be extended accordingly. The case of the part enables to test the electrospinning process in vertical or horizontal position. In order to provide optimal condition for electrospinning, the pulley parts and the dosing cartridge and guide part is covered by covers *7*. The active electrospinning zone in between these covers is then *150 mm* wide.

3. THEORETICAL ANALYSIS

In order to predict the behaviour of the designed electrospinning device, the electrostatic analysis was carried out. For this purpose, the software Autodesk Simulation Mechanical was used. This tool enables to calculate the electric potential and electric intensity distribution over the parts in the model using the finite element method. The 2-D cross section of the analysed model of the wire spinner is shown in the **Fig. 2a**. This model consists of the wire spinner covered by circular parts representing the polymer films, the plate collector and the outer space describing the surrounding air. Material properties are defined by the dielectric constant ε_r, see the **Table 1**. Selected parts of the model are subjected to appropriate boundary condition definition: the positive electrical potential on the surface of the wire part, the negative electrical potential on the surface of the collector part and the zero electrical potential on the outer boundary of the air part. The values of defined voltage on each electrode during the analysis are listed in the **Table 1**.

Table 1 Boundary condition definition and material properties of the model parts

Part of the model	Voltage [kV]	Dielectric constant [-]	Size [mm]
Wire (steel)	-15	10^6	Diameter D_w=0.48
Collector (steel)	15	10^6	Rectangle 150x10
Outer boundary of air	0	1	Square 450x450
Polymer - core	-	20	D_c= 0.53
Polymer - shell	-	20	D_s=0.67
Electrodes distance	-	-	H=150

Results of the electrostatic analysis of the 2-dimensional model of the wire spinneret are shown in the **Fig. 2**. The **Fig. 2b** shows the electric field distribution and the **Fig. 2c** describes the distribution of the electrical field strength E.

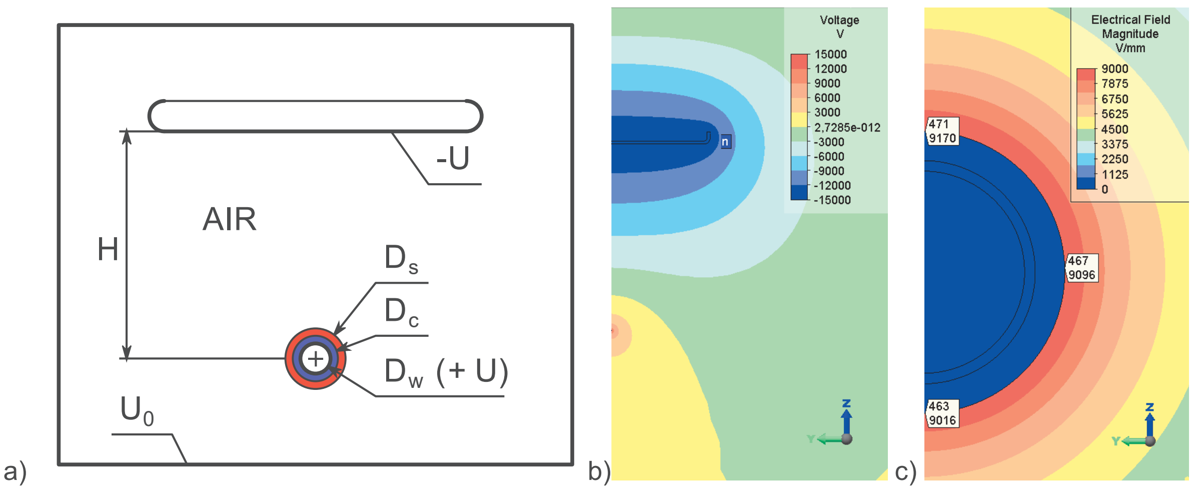

Fig. 2 a) The 2-D wire spinner model layout in the software environment Autodesk Simulation Mechanical. **b)** Results: Voltage distribution (half-model) **c)** Results: Electric field strength (detail of the wire electrode)

According to theory of electrospinning, the condition for the start of the electrospinning process is a reaching of the critical electrical field strength E_c on the polymer surface. In the electrostatic problem, the electrical intensity is a function of the surface curvature. However, according to the theory of wave [9], this is further affected by wavelength. Results of the electrostatic problems are therefore approximate approach to the problem, neglecting the dynamic effects of the solution in the electric field.

Using the described model, the influence of various parameters on the electric intensity E was analysed. Results of these analyses are shown in the graphs in the **Fig. 3**.

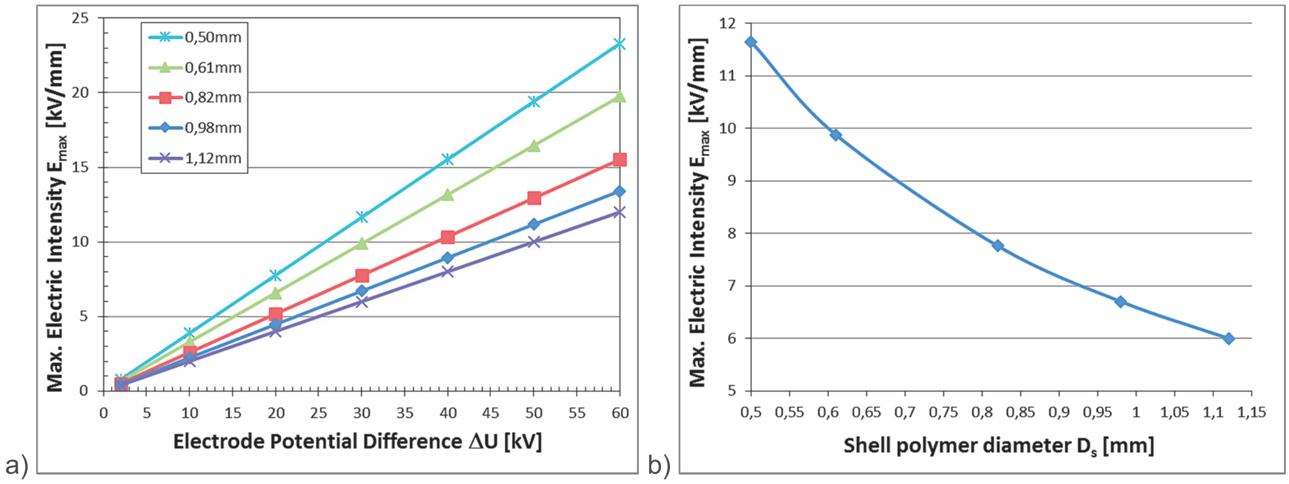

Fig. 3 Results of the electrostatic analysis **a)** Dependency of Max. Electric Intensity E_{max} on the electrode potential difference ΔU (under constant electrode distance H=150 mm) **b)** Dependency of Max. Electric Intensity E_{max} on the shell polymer diameter D_s (H=150 mm, ΔU=30 kV)

4. EXPERIMENT

Experiments were carried out using the above described manufactured testing device. In preliminary tests, only the shell polymer was applied in order to set the optimal winding and dosing parameters. In the next step, experiments with the polymer two-layer composed of core and shell polymer solution were carried out. These experiments were based on results obtained from initial non-coaxial tests. The critical value of voltage U_c, the critical value of the electric field strength E_c and the distance between adjacent polymer jets corresponding with the characteristic wavelength λ were investigated during our experiments.

Material used for our experiments was a water-soluble Polyvinyl alcohol (PVA, Sloviol R16, Chemicke zavody Novaky, SK). For the shell material was prepared a solution of 12 wt.% PVA in DI water. The solution of 4 wt. % PVA dissolved in DI water was prepared as the core material. The Prussian Blue $[Fe_4[Fe(CN)_6]_3]$ was added to the core solution for better observation of coaxial electrospinning and for an analyse of the core/shell structure. The wire electrode was connected to a positively charged (*+ 35 kV*) a high voltage source (Spellman Sl100). The plate collector was connected to the negatively charged (*-30 kV*) high voltage source (Spellman Sl100). The optimal winding speed of the wire for maintaining of a homogeneous polymer two-layer and a complete electrospinning of polymer solutions was *7 mm/s*. Polymer solutions were dosed at constant rate of 9 ml/h (the shell solution) and of *3 ml/h* (the core solution). The electrospinning was carried out at ambient temperature 23°C and at a relative humidity *31 %*. Core/shell nanofibers were collected on the Spunbond nonwoven (Pegas Nonwovens, CZ), located under the plate collector.

For the detail observation of the process of the polymer solution application and also the Taylor cone and polymeric jet formation, the high speed camera Olympus i-SPEED 3 was used. The track of Taylor cone and double polymer jet formation was recorded at the rate 2000 and 5000 frame per second with the image resolution 1280x1024, resp. 804x600 pixels. For this purpose, the ultra-high performance light source Olympus ILP-1 was used. The image from the high speed camera is shown in the **Fig. 4a**. The image in the **Fig. 4b** shows the active zone of wire spinner recorded by an HD camera.

There was a significant reduction of the feed rate in case of coaxial electrospinning. The reason was, that it is more demanding to maintain the uniform polymer two-layer throughout the coaxial electrospinning process. A sensitive dosing ensures easier process control. Many polymer jets were observed during coaxial electrospinning process. Polymer jets occurred throughout the length of the wire electrode and over its entire perimeter, see (**Fig. 4 a, b**).

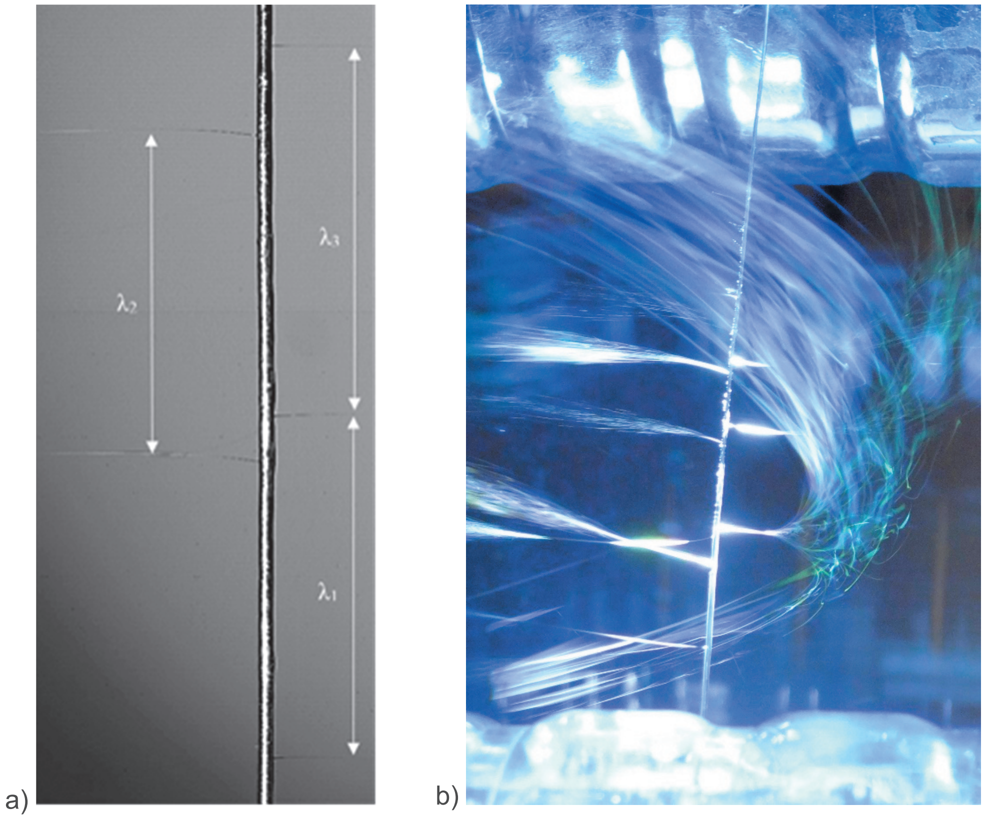

Fig. 4 Experiments results

a) An image from high speed camera with highlighted wavelength λ

b) An image of wire spinner (undisclosed plate collector is placed at the left vertical side of the picture) from HD camera

5. DISCUSSION

Experiments verified that the optimal configuration of this wire spinner for coaxial electrospinning is in the vertical direction, were the wire is wounded from the bottom pulley upwards vertically. In this configuration it is possible to apply the double polymer layer onto the wire electrode. The winding speed and the amount of polymers dosed into the feeding chambers are crucial parameters for setting up the optimal condition for coaxial electrospinning. With the aim to produce as thin polymeric film as possible, it was observed that very thin films are more likely to be turned into droplets when the high voltage is applied. This may reflect to the theory of Rayleigh instability. The average value of λ, evaluated from the high speed camera track is 13 mm. However, seldom jets with a lover value occurred in the track too. Polymer jets were formed on the wire layer in all direction (not only in the direction towards the collector as was expected). This effect (clearly seen in the **Fig. 4**b) may relate to relatively equal value of intensity over the whole perimeter of the polymer on the electrode. This phenomenon was present also in other experiments, where the metal wire was replaced by PA monofilament of the same diameter. Due to the poor conductive properties of this material, the electric charge was distributed over the electrode through the polymer layer only. The stability and number of polymeric jets in the active zone remained approximately equal to the tests with a metal wire. Reaching the critical value of electric intensity E_c on the polymer surface closer to collector can be achieved by a proper setting of the electrodes voltage difference. According to analysis results (see **Fig. 3**a), the maximal electric intensity on the polymer surface depends linearly on the applied voltage.

The Morphology of a produced nanofibrous layer was observed using scanning electron microscope (SEM) Tescan Vega 3 SB (Tescan, CZ).The core/shell structure of produced nanofibers was investigated using SEM Phenom G2, (FEI, USA). The diameters of nanofibers were measured with the image analysis software NIS elements 3.0. The average diameter values of nanofibers was 570,86±215 nm.

6. CONCLUSION

The presented work illustrates complete analysis of coaxial electrospinning process related to a wire spinner. It describes the design of wire spinneret supplemented with the analysis of electrostatic field and strength of corresponding system. Carried out experiments proved the ability of application of double polymer layer on the wire electrode, that is considered as an important precondition for the coaxial electrospinning process. It can be concluded, that the presented system enables creating of homogenous double polymer film that can be then electrospun into core/shell fibers and collected on the collector. However, the optimal dosing and winding parameters requires sensitive setting defined carefully for particular polymers. Despite the complexity of proof of the core/shell structure of nanofibers, the analysis of the nanofibers suggests the core/shell nanofibers in the material from performed experiments.

Thanks to its design, a wire spinner technology may find its potential in industrial sector. However, further research regarding an increase of productivity of presented technology may be beneficial. Application of the wire spinner for an AC electrospinning and further analysis of possible method for proof of the core/shell nanofibrous structure will be therefore the subject of upcoming research in this field.

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REFERENCES

[1] TONG L. et al.: Nanofibers - Production, Properties and Functional Applications. : InTech, Chapters published November 14, 2011 under CC BY 3.0 license, ISBN 978-953-307-420-7,DOI: 10.5772/916.

[2] JIRSAK, O., et al.: A method of nanofibers production from polymeric solution using electrostatic spinning and device for carrying out the method. Technical University of Liberec. WO2005024101 (A1). 2015.

[3] POKORNY, P., et al. (2014). Effective AC needleless and collectorless electrospinning for yarn production. Physical Chemistry and Chemical Physics (PCCP), Royal Society of Chemistry, Cambridge. DOI: 10.1039/c4cp04346d.

[4] MALY, M., et al.: Method for application of liquid polymeric material onto spinning cords and a device for production of nanofibers through electrostatic spinning. ELMARCO SRO, WO2012139533.

[5] REZNIK, S. N., et al. (2006). Evolution of a compound droplet attached to a core-shell nozzle under the action of a strong electric field. Phys. Fluids, 18 (6).

[6] POKORNY, P., et al.: Weir spinner, Czech Republic Patent PV 2009-425, 2009.

[7] FORWARD, K.M. et al. (2013). Production of core/shell fibers by electrospinning from a free surface. Chemical Engineering Science. , pp. 250-259.

[8] HOLOPAINEN, J. et al. (2014). Needleless electrospinning with twisted wire spinneret, Nanotechnology 26 025301 doi:10.1088/0957-4484/26/2/025301.

[9] LUKAS, D. et al. (2008). Self-organization of jets in electrospinning from free liquid surface: A generalized approach, Journal of Applied Physics, Vol. 103, No. 8, 0843091-7.