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ELECTROSPUN BIOACTIVE POLYMER/GELATIN COATINGS FOR MEDICAL APPLICATION - CREATION AND INVESTIGATION

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ABSTRACT

Skin protects human body from negative external influences; however, it has property to be injured. There are a lot of different types of medical dressings, nevertheless, electrospun materials for medical applications are on demand due to nonwoven materials properties, such as large surface area, specific porosity, polymers suitability. Natural polymers for wound dressings are highly investigated not only for their antimicrobial and anti-inflammatory properties, but for electrospinning feasibility as well. The aim of this research is to develop bioactive polymer/gelatin coating from animal waste and investigate its feasibility for medical dressing. Polyvinyl butyral (PVB) and polyvinylpyrrolidone (PVP) as polymers were chosen due to their adhesive properties, solubility in various solvents and biocompatibility. Gelatin as bioactive polymer was chosen due to its anti-inflammatory properties. Natural *Symphytum officinale* (SO) extract in compositions was used. Electrospun polymeric nano/microfibers were coated with gelatin compositions. Nano/microfibers diameters and gelatin compositions droplets sizes were evaluated. It was determined that composition modification with SO has influence on used polymers nano/microfibers diameter. Plant extract addition to polymer solution induces thinner nano/microfibers. Consequently, it was ascertained that gelatin and SO composition forms high quality droplets both on PVB/SO and PVP/SO nanofibers surface.

KEYWORDS

Electrospinning, nano/microfibers, polyvinyl butyral, polyvinylpyrrolidone, gelatin, *Symphytum officinale*.

INTRODUCTION

The skin as the largest human organ protects internal organs and tissues from physical, chemical, and/or biological influences. However, skin has property to be injured, therefore fast regeneration of skin tissues and restoring security barrier is obligatory [1]. Nonetheless, wound healing is a very complex process when unsuccessful treatment may have objectionable consequences on human life. To facilitate and accelerate wound healing different types of medical dressing are used. Bioactive dressings are made from natural or artificial biomaterials (collagen and/or alginate) and ensure biocompatibility and non-toxicity. Bioactive dressings (collagen) initiate the synthesis of fibroblasts and accelerate endothelial migration, what determines more rapid wound healing [2].

A lot of miscellaneous types of medical dressings are used – films, hydrogels, foams, hydrocolloids, nanoparticles, etc. Tissue engineers still pay high attention to natural intercellular matrix investigation and imitation creation to produce medical dressings. Therefore, electric spinning as one of leading technologies is highly on demand due to feasibility to form a nonwoven fabric from polymer solution.



Nonwoven fabric can absorb extensive amount of wound secreted exudate and protect wound from dehydration as well due to large specific surface area. High porosity surface ensures air and moisture permeability and protects wound from microorganisms. Furthermore, the feasibility to use different combinations of polymers solutions is one of the main advantages of this method [3].

Natural polymers extracted from plants, animals or microorganisms are desirable for the electric spinning process due to their practical application, durability, chemical and physical stability and low antigenicity. Collagen, gelatin, and/or hyaluronic acid have antimicrobial and anti-inflammatory properties, therefore are particularly valued in the medical industry [4].

The aim of this research is to develop bioactive polymer/gelatin coatings from animal waste and investigate its feasibility for medical dressings.

MATERIALS AND METHODS

Polyvinyl butyral (PVB), Mowital B60T from Kuraray, and polyvinylpyrrolidone (PVP), $M_w=1300\ 000$ LS purchased from Sigma Aldrich, Germany, as carrier polymers for electrospinning were used. The gelatin (GEL) with the high concentration of collage of type I-III (collagen protein content 71.5%) obtained from the anatomical parts of organically reared bulls was obtained from MB Kulagenas. Comfrey (*Symphytum officinale* or *S. officinale*, SO) root extract in ethanol (C=10%) was prepared by Lithuanian Research Centre for Agriculture and Forestry and uses as bioactive additive in polymer compositions.

Polymer solutions were prepared by dissolution of polymers in ethanol (96.6%) by constant stirring (PVP – 3 h, PVB – 24 h) with magnetic stirrer to obtain homogeneous solutions. PVB and PVP compositions used are listed in Table 1.

Table 1. Polymer compositions.

| | PVB, % | PVP, % | GEL, % | SO, % | ETHANOL, % | DISTILLED WATER, % |
|---------------|-----------|-----------|-----------|----------|---------------|-----------------------|
| PVB | 10 | | | | 90 | |
| PVB/SO | 10 | | | 2 | 88 | |
| GEL | | | 20 | | 48 | 32 |
| GEL/SO | | | 20 | 2 | 47 | 31 |
| PVP | | 10 | | | 90 | |
| PVP/SO | | 10 | | 1 | 89 | |

Electrospun micro/nanofiber mats were formed using electrospinning machine „Nanospider™“ (Elmarco, Check Republic) equipped with rotating spinneret (distance between electrodes 130 mm) at ambient conditions. The operating voltage was 35–45 kV and 40–50 kV for PVB and PVP, respectively (Table 2).

Table 2. Electrospinning (electrospraying) settings.

| | VOLTAGE, kV | SPINNING TIME, MIN | | VOLTAGE, kV | SPINNING TIME, MIN |
|-------------------|----------------|-----------------------|-------------------|----------------|-----------------------|
| PVB(35) | 35 | 4 | PVP(40) | 40 | 3 |
| PVB(45) | 45 | 4 | PVP(50) | 50 | 3 |
| PVB/SO(35) | 35 | 4 | PVP/SO(40) | 40 | 3 |
| PVB/SO(45) | 45 | 4 | PVP/SO(50) | 50 | 3 |

GEL and GEL/SO solutions (C=20%) were heated to 30-35 °C before electrospinning on electrospun PVB and PVP mats at 55 kV voltage for 10 min.

The structure of micro/nanofibers were analysed by scanning electron microscope S-3400N (SEM). The diameters of 100 micro/nanofibers were calculated using Nis-Elements D 4.50.00 (Nikon) from SEM images.

RESULTS AND DISCUSSION

SEM images and diameter distribution histograms of electrospun PVB, PVB/SO PVP, PVP/SO mats are presented in figure 1. As can be seen, diameter of PVB and PVB/SO micro/nanofibers increase as operating voltage increases (diameter range of 131 nm – 915 nm at 35 kV and 279 nm – 1757 nm at 45 kV). The addition of *S. officinale* extract slightly reduces PVB/SO micro/nanofibers diameter (to the range of 292–856 nm at 35 kV and 406–1091 nm at 45 kV). PVP micro/nanofibers diameter is almost independent on voltage (282–1216 nm at 40 kV, 263–1187 nm at 50 kV). *S. officinale* extract also decreases diameter of PVP/SO micro/nanofibers leading to larger number of micro/nanofibers with tiny diameter.

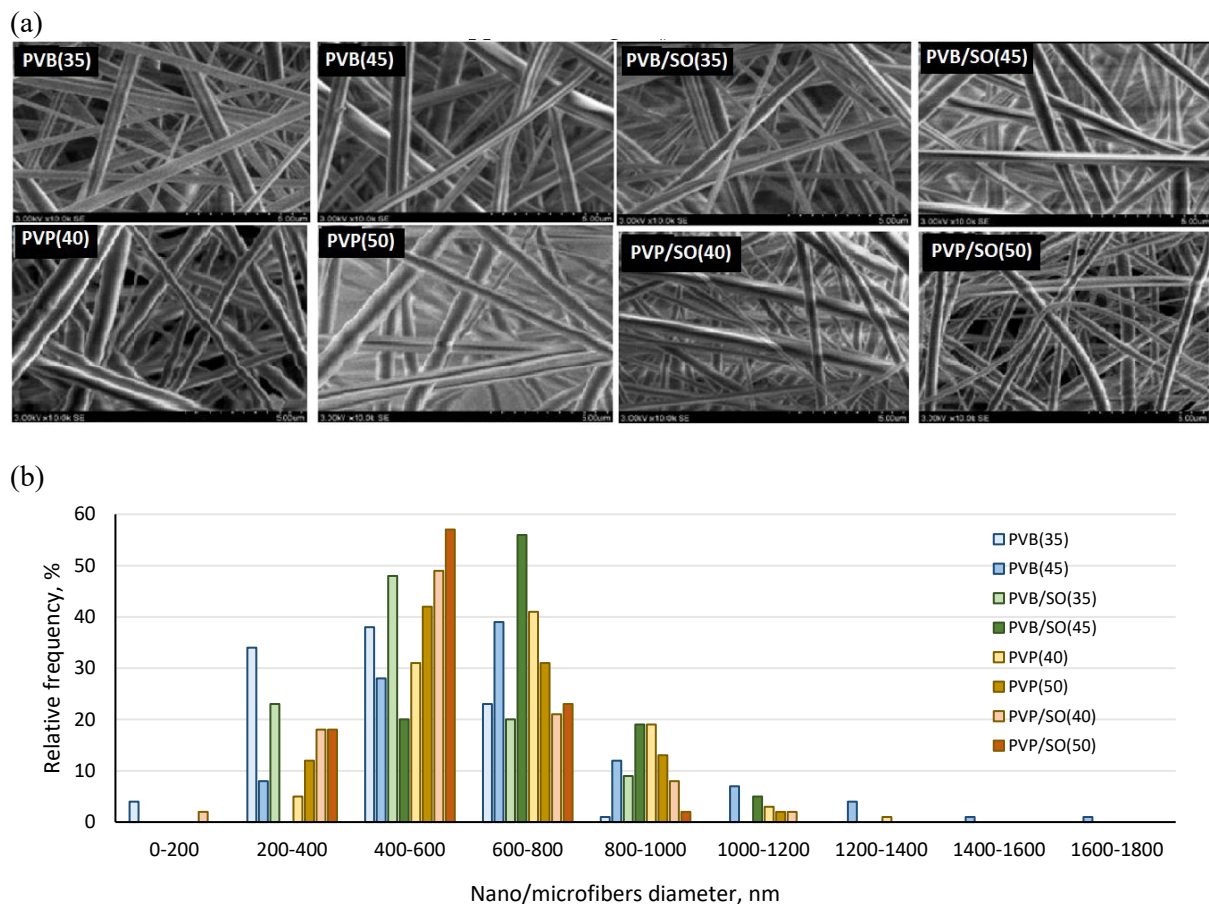


Figure 1. Polymer composition and operating voltage influence on micro/nanofibers diameter distribution: a – SEM images, b – histograms.

In this study attempts were also made to obtain a gelatin mat by electrospinning. However, gelatin exhibits poor electrospinnability due to hydrogen bonding [5]. Therefore, it was not possible to obtain nanofibers from gelatin solution. Nevertheless, during electrospinning gelatin spraying process take place and droplets were formed on PVB and PVP micro/nanofiber mats (Fig. 2). As in the case of PVB and PVP micro/nanofibers, the droplets size of gelatin also depends on operating voltage – increase of voltage value results on the increase of droplets main diameter.

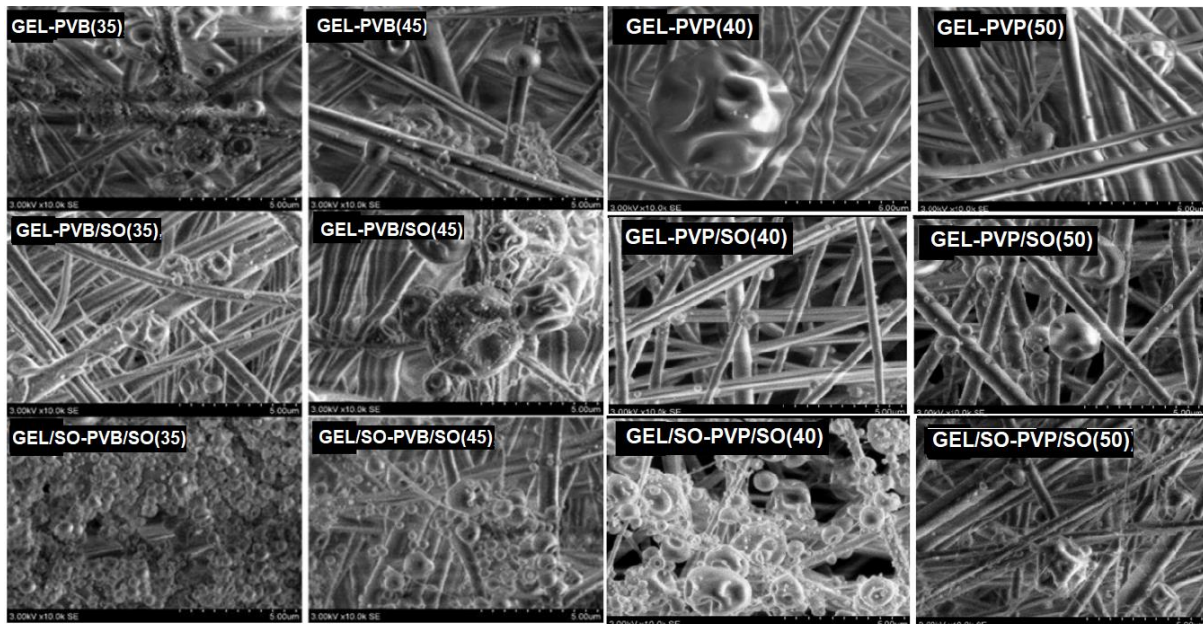


Figure 2. Composition and electrospinning voltage influence on gelatin droplets quality (SEM pictures).

From Fig. 2 is evident, addition of *S. officinale* extract into both polymer and gelatin compositions increase droplets density on the PVB/SO and PVP/SO micro/nanofibers.

CONCLUSION

Addition of bioactive *Symphytum officinale* extract to PVB and PVP compositions lead to the formation higher number of micro/nanofibers with smaller diameter.

Due to poor electrospinnability of gelatin only gelatin droplets were sprayed on PVB and PVP micro/nanofibers mats. The density of gelatin droplets formation can be increased by the addition of bioactive *Symphytum officinale* extract.

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