Production and Characterization of Poly(vinyl alcohol)/Poly(vinylpyrrolidone) Iodine/Poly(ethylene glycol) Electrospun Fibers with (Hydroxypropyl)methyl Cellulose and Aloe Vera as Promising Material for Wound Dressing

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ABSTRACT: Biocompatible poly(vinyl alcohol)/poly (vinylpyrrolidone) iodine/poly(ethylene glycol) fibers containing (hydroxypropyl)methyl cellulose (HPMC) and aloe vera were successfully prepared by electrospinning their aqueous solution. Aloe vera which is known to be effective in the treatment of various wounds was added to the polymer solution. HPMC was added to the system as the water retention agent. The hybrid fiber mats were subjected to detailed analysis using a differential scanning calorimeter, a scanning electron microscope (SEM), and a Fourier transform infrared spectrometer. Images obtained from the SEM showed that the polymer fibers were linear, homogenous, and contained no beading. The fiber diameters ranged between 100 and 900 nm. It was seen that the electrospun mats obtained could potentially be used as a material for dressing wounds. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 124: 3520–3524, 2012

Key words: fibers; polymer blends; copolymers; biocompatibility; biodegradable

INTRODUCTION

This study aims to produce a wound dressing material especially for burns by electrospinning poly (vinyl alcohol) (PVA), poly(vinylpyrrolidone) iodine (povidone iodine) (PVP-I), and a poly(ethylene glycol) (PEG) hybrid polymer containing aloe vera and (hydroxypropyl)methyl cellulose (HPMC). Aloe vera is a plant, used in the treatment of burns, similar to propolis, marigold, and so forth. According to chemical data, aloe vera contains various carbohydrate polymers, notably glucomannans, along with a number of other organic and inorganic components.[1,2] Many of the health benefits associated with aloe vera, especially in relationship with healing wounds, have been attributed to the polysaccharides contained in the gel in its leaves.[3–8]

PVA has a number of desirable characteristics that makes it a good material for dressing burns. PVA has sufficient mechanical strength and high flexibility, and water swells it.[9]

PEG can be used on burns, and it is nontoxic, biocompatible, and hydrophilic. PEG has good mechanical properties and high levels of hydration which make it a very suitable candidate for a wound dressing material.[10]

Poly(vinylpyrrolidone) (PVP) is a synthetic polymer with good biocompatibility. PVP bound iodine (PVP-I) was developed by NASA for the Apollo program and first used in the Apollo 11 space journey in 1969. Its use in medicine followed soon after. PVP-I is beneficial on burn wounds due to its effect on the reduction of bacterial colony counts.[11]

In this study, aloe vera was blended with HPMC to increase water retention properties of the end material. HPMC was mainly used as thickener, stabilizer, emulsifier, excipient, water retention agent, film-forming agent, and so on which are very important for the preparation of wound dressing materials.

The chemical structure of HPMC is given in Figure 1.

The wound dressing fibers were obtained by the electrospinning technique in this study. Electrospinning is a cost effective way of manufacturing wound dressing materials. The electrospun fiber mats show controlled evaporative water loss, excellent oxygen permeability, good fluid drainage ability, and inhibit exogenous microorganism invasion because of its ultrafine pores. Histological examination indicates

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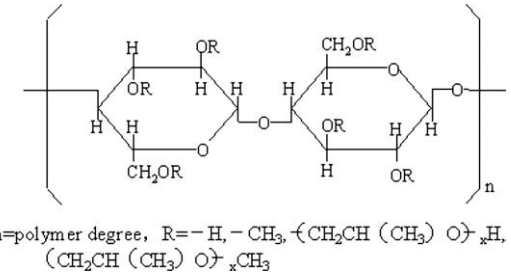

Figure 1 Chemical structure of HPMC.

that the rate of epithelialization is increased, and the dermis becomes well organized, if wounds are covered with electrospun fiber mat. This electrospun mat can be potentially used for wound dressings because of its unique properties.[12]

EXPERIMENTAL PART

Materials and methods

PVA (MW: 85,000–124,000 g mol^{-1}, 98% hydrolyzed) and PEG (Merck, MW 400) (MW 400,000 g mol^{-1}) were purchased from Merck (Darmstadt, Germany). Granular PVP-I and HPMC powder were supplied by BASF (Ludwigshafen, Germany). The aloe vera gel solution (50 mL) was supplied by Forever Living Health and Beauty Products (Scottsdale, Arizona). Ultrapure water produced from an MPI system was used for all the experiments.

The experimental procedure of this study consisted of three major steps: (i) the preparation of the hybrid polymer solution. (ii) The electrospinning of the hybrid polymer solution to generate PVA/PVP-I/PEG fibers containing HPMC and aloe vera. The spinning experiments are usually performed at the room temperature. (iii) Characterization of obtained fibers by differential scanning calorimeter (DSC), Fourier transform infrared spectrometer (FTIR), and scanning electron microscope (SEM).

Preparation of the hybrid polymer solutions

PVA hydrogel was prepared by fully dissolving with magnetic stirring 10 g of polymer powder in the corresponding amount of ultrapure deionized water for 2 h, at the temperature of 80 ± 2°C. PVA solution (10%) was cooled down to room temperature. Solution A was prepared by mixing 100 g PVA, 10 g PVP-I (10 wt %), 1 g PEG (10 wt %), and 2 g HPMC at 60°C with magnetic stirring for 2 h. Solution B was obtained by adding 2 g of aloe vera (2 wt %) to Solution A. Solution C was prepared by adding 4 g of aloe vera (2 wt %) to Solution A. Finally, Solution D was prepared by adding 6 g of aloe vera (2 wt %) to Solution A.

Electrospinning of the hybrid polymer solutions

Polymeric fibers can easily be prepared by the electrospinning technique. The electrospinning process is based on the use of a high electric field to draw a polymer solution from the tip of a capillary toward a collector. The voltage of the electrospinning equipment is adjusted with variable high-voltage power supply from Gamma High Voltage Research. Fiber samples were prepared by the electrospinning of four different aqueous solutions at an electrical voltage of 20 kV at room temperature under atmospheric pressure. The applied voltage causes a jet of the solution to be drawn toward a grounded collector. The syringe needle (8 mm) was used as the electrode connected to the power source. Fine jets form nanosized polymeric fibers, after they dry up and they are collected as a web-like mass. The collector was a 20 × 40 cm^2 aluminum foil placed horizontally 15 cm away from the tip of the needle. The collector was connected to the power supply as an electrode with opposite polarity. A metering syringe pump from New Era Pump Systems was supplied the polymer solution at a constant rate of 0.5 mL h^{-1}. Finally, fibers were detached from Al foil collector and dried in the furnace at 70°C overnight under vacuum.

Measurement and characterization

Formation and morphology of the electrospun PVA/PVP-I/PEG with HPMC and aloe vera fibers were determined using a SEM Quanto 400 FEI MK-2.

FTIR with Attenuated Total Reflectance (ATR) module was obtained using a Thermo Nicolette 6700 spectrophotometer. DSC studies of the fibers were carried out using a Schimadzu DSC-60 DSC.

pH and conductivity of the hybrid polymer solutions were measured using a Wissenschaftlich-Technische-Werkstätten and a 315i/SET apparatus, and the viscosity of the hybrid polymer solutions were determined with an AND SV-10 viscometer.

Fiber diameters were quantitatively measured using the ImageJ software. ImageJ is a Java-based public domain program that contains basic digital image processing tools and includes numerous tools that facilitate quantitative measurements which was originally developed at the National Institute of Health.[13]

RESULTS AND DISCUSSION

Physical properties of the hybrid polymer solutions and fibers

Table I lists the pH, viscosity, conductivity, and surface tension values of the hybrid polymer solutions.

TABLE I
Physical Properties of the Hybrid Polymer Solutions

Sample name	Viscosity of solution (mPa s)	Conductivity of solution (μS cm^{-1})	pH	Surface tension (mN m^{-1})
Solution A	461	1161	5.18	74
Solution B	523	1040	5.34	76
Solution C	603	928	5.90	77
Solution D	610	698	5.95	79

Addition of aloe vera increased the pH of the hybrid polymer solutions.

The addition of aloe vera from 2 to 6% decreased the conductivity of the solution from 1040 to 698 μS cm^{-1} but increased the viscosity and surface tension of the solution from 523 to 610 mPa s and 76 to 79 mN m^{-1}, respectively.

DSC analysis of the fibers

DSC measurements were carried out using nitrogen as the carrier gas. The temperature was raised from room temperature to 200°C, then it was cooled down to room temperature, and the sample was heated again to 500°C at a rate of 10°C min^{-1}.

The DSC thermograms of the fibers obtained from Solutions B, C, and D are given in Figure 2. As seen from Figure 2, the increase in amount of aloe vera caused a slight shift in the melting points and decreased the intensity of the resulting peak. However, the increase in amount of aloe vera caused broadening of resulting peak. It could be easily seen from thermogram c in Figure 2 that there is a broad peak indicates glass transition temperature instead of melting point. This indicates that the increase in amount of aloe vera affects the crystal structure and efficiently crosslinks the hybrid polymer forming an amorphous structure.

Figure 2 shows that the increase in amount of aloe vera caused a positive shift in both the starting temperature of the decomposition process and the temperature where the highest decomposition occurs. The decrease in the size of the decomposition with the increase in amount of aloe vera is indicative of a more thermally strong structure with an increasing degree of crosslinking. It is clear that the increase in amount of aloe vera increased the thermal strength of the material.

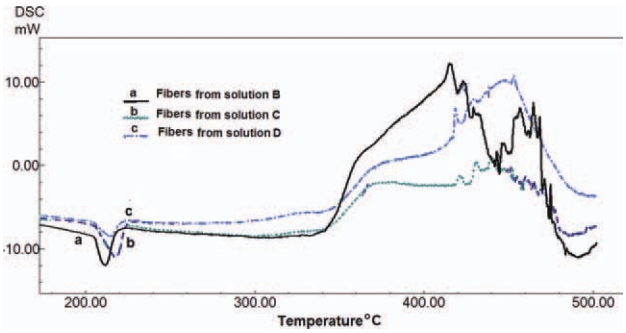

Figure 2 The DSC thermograms of the fibers obtained from Solutions B, C, and D containing 2, 4, and 6% aloe vera, respectively. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

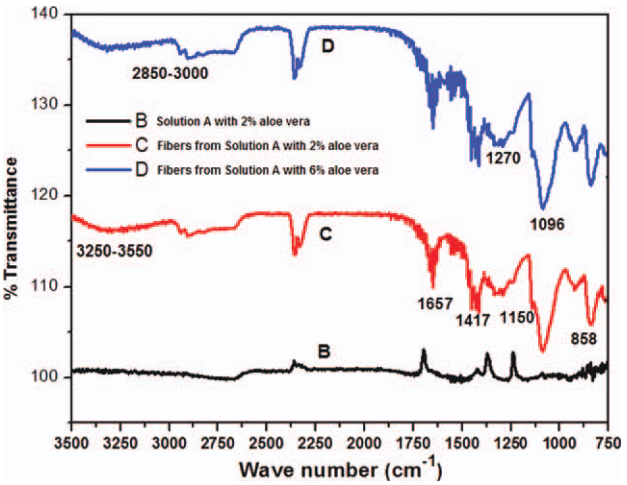

Figure 3 FTIR spectra of the Solution B containing 2% aloe vera, its electrospun fibers, and electrospun fibers of Solution D containing 6% aloe vera. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

FTIR analysis

Figure 3 is the FTIR spectra of the PVA/PVP hydrogels obtained within the range between 3500 and 750 cm^{-1}. The absorption peaks at 1417 cm^{-1} (CH_2 bending) and 858 cm^{-1} (CH_2 rocking) are characteristic to PVA appear in all spectra. However, the peak heights are different. The peak at 1270 cm^{-1} is due to the C—H vibration. The broad peak at 1096 cm^{-1} indicates the C—O stretching vibration. The absorption peak at 1657 cm^{-1} is characteristic C=O group and is present in all spectra. From Figure 3, C—H broad alkyl stretching band (2850–3000 cm^{-1}) and hydrogen bonded band (3250–3550 cm^{-1}) can be clearly seen. Intramolecular and intermolecular hydrogen bonding is expected to occur among PVA chains due to high hydrophilic forces. These peaks are absent for the PVA electrospun fibers as expected. The absorption peak at 1096 cm^{-1} (C—O, 1090–1150 cm^{-1}) is very sharp for the electrospun fiber. This is a carboxyl stretching band (C—O) and is attributed to the crystallinity of the PVA. It is used for the assessment of PVA structure.

SEM analysis

The ImageJ digital image analysis software was used to measure the fiber diameters.[13] The fibers obtained

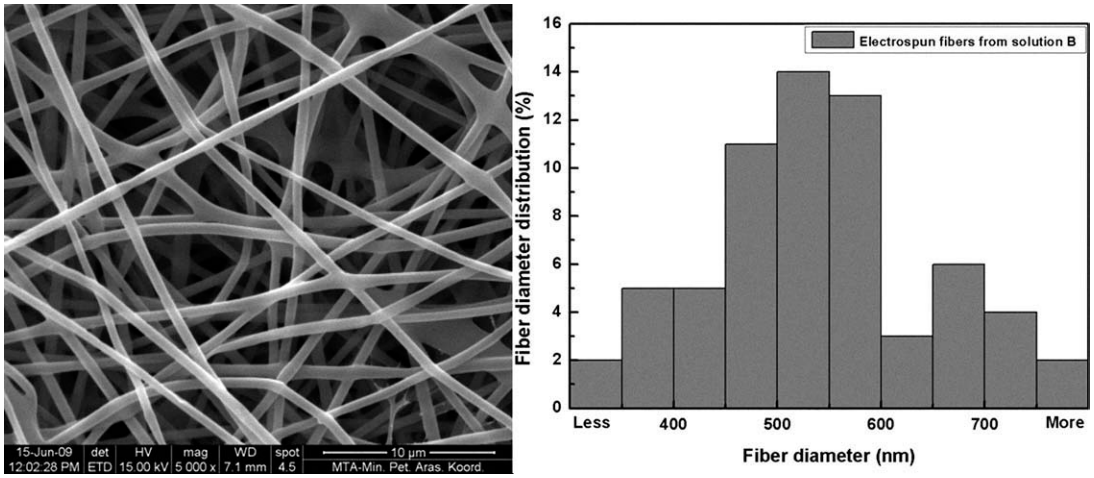

Figure 4 (a) SEM picture of the electrospun fibers obtained from Solution B containing 2% aloe vera. (b) Fiber diameter distribution graph of the electrospun fibers obtained from Solution B containing 2% aloe vera.

from Solution B containing 2% aloe vera are given in Figure 4(a). The average diameter of the fibers is 546 nm. It is clear that the fibers are linear and contain no beading. Fiber size distribution of PVA/PVP-I/PEG containing HPMC and 2% aloe vera (Solution B) electrospun fiber mats is presented in Figure 4(b). Diameters of electrospun PVA/PVP-I/PEG fibers containing HPMC and 2% aloe vera range from 0 to 900 nm. The electrospun fibers obtained from Solution C are given in Figure 5(a). The average diameter of the hybrid polymeric fibers obtained from Solution C containing 4% aloe vera was calculated as 416 nm. It is clearly seen that the fibers obtained from Solution C are linear and contain no beading. Fiber size distribution of PVA/PVP-I/PEG containing HPMC and 4% aloe vera (Solution C) electrospun fiber mats is presented in Figure 5(b). Diameters of the electrospun PVA/PVP-I/PEG fibers containing HPMC and 4% aloe vera are slightly finer and range from 200 to 750 nm. Figure 6(a) shows the electrospun fibers obtained from Solution D containing 6% aloe vera. The average diameter of the hybrid polymeric fibers obtained from Solution D containing 6% aloe vera was calculated as 278 nm. It also does not contain beading as the other fibers. Fiber size distribution of PVA/PVP-I/PEG containing HPMC and 6% aloe vera (Solution D) electrospun fiber mats is presented in Figure 6(b). Diameters of the electrospun PVA/PVP-I/PEG fibers containing HPMC and 6% aloe vera range from 100 to 550 nm. It could be easily seen from fiber diameter distribution graphs that the most of electrospun PVA/PVP-I/PEG fibers containing HPMC and 6% aloe vera range from 150 to 350 nm, the most of electrospun PVA/PVP-I/PEG fibers containing HPMC and 4% aloe vera range from 250 to 500 nm, and the most of electrospun PVA/PVP-I/PEG fibers containing HPMC and 2% aloe vera range from 350 to 700 nm. SEM results

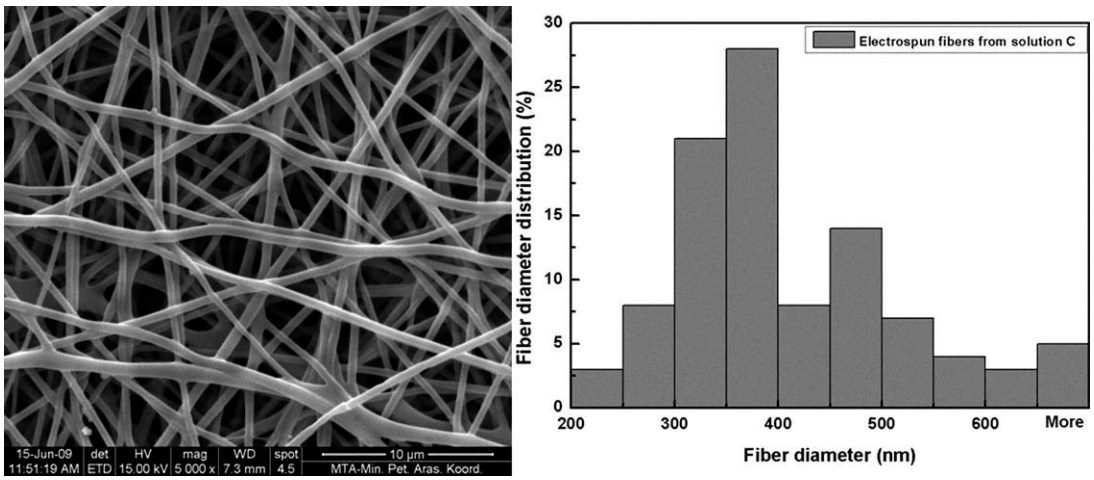

Figure 5 (a) SEM picture of the electrospun fibers obtained from Solution C containing 4% aloe vera. (b) Fiber diameter distribution graph of the electrospun fibers obtained from Solution C containing 4% aloe vera.

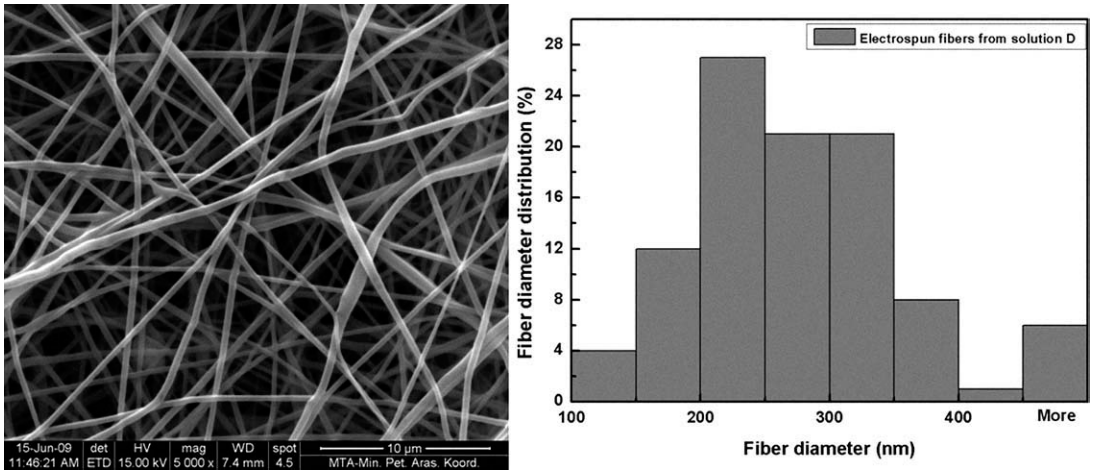

Figure 6 (a) SEM picture of the electrospun fibers obtained from Solution D containing 6% aloe vera. (b) Fiber diameter distribution graph of the electrospun fibers obtained from Solution D containing 6% aloe vera.

and fiber diameter distribution graphs clearly show that the electrospun PVA/PVP-I/PEG fibers containing HPMC and 6% aloe vera are finer and more homogenous with respect to other fibers.

CONCLUSIONS

PVA/PVP-I/PEG electrospun fibers were fabricated following the addition of aloe vera and HPMC. The DSC results show that the increase in amount of aloe vera shifts the melting point of the polymers to higher temperatures, and the increase in amount of aloe vera causes a positive shift in both the starting temperature of the decomposition process and the temperature where the highest decomposition occurs. Amorphous structure was conformed instead of crystal structure due to the increase in amount of aloe vera. Fibers obtained from Solution D have glass transition temperature higher than the melting points of other fibers. This indicates that the thermal stability of the resulting fibers increased. The FTIR results were also in good accordance with data in the literature and characterize the main structure of the PVA/PVP-I/PEG polymer hybrid. The SEM micrographs show that the increase in amount aloe vera results in the formation in finer fibers without any beading. This increases the porosity and facilitates the penetration of oxygen and moisture to the wound, which are of vital importance for the healing process. The increased porosity of the structure effectively prevents the infiltration of bacteria. The fibers prepared with 6% aloe vera are much more porous, which makes them highly suited for use in wound dressing materials.

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