

Simply Electrospun Gelatin/Cellulose Acetate Nanofibers and their Physico-Chemical Characteristics

1 Simply Electrospun Gelatin/Cellulose Acetate Nanofibers and their Physico-Chemical Characteristics

Fuji Hernawati Kusumah^{1,3}, Ida Sriyanti^{1,3}, Dhewa Edikresnha^{1,3},
Muhammad Miftahul Munir^{2,3} and Khairurrijal^{1,3,a}

¹Physics of Electronic Materials Research Division,

²Theoretical High Energy Physics and Instrumentation Research Division,

³Bioscience and Biotechnology Research Center,

Institut Teknologi Bandung

Jalan Ganesa 10, Bandung 40132, Indonesia

^akrijal@fi.itb.ac.id

Keywords: Electrospinning, gelatin, cellulose acetate

Abstract. Gelatin in fibers form can be used for tissue engineering, wound dressing, or drug carrier. However, it is easily damaged if exposed to water. Thus, it was blended with cellulose acetate. Acetic acid was used as a solvent because it is less toxic. The mass ratios of gelatin to cellulose acetate of 10:0, 8:2, and 6:4 were as precursor solutions. Simple electrospinning was employed to produce gelatin/cellulose acetate fibers. From SEM images, it was shown that the average diameters of gelatin/cellulose acetate fibers from the precursor solutions of 10:0, 8:2, and 6:4 were 534, 649, and 765 nm, respectively. The addition of cellulose acetate increased the viscosity of gelatin/cellulose acetate solution. Moreover, gelatin mass reduction caused a decrease in conductivity of gelatin/cellulose acetate solution. Therefore, increasing in the viscosity or reducing in the conductivity of the precursor solution increased the average diameter of the gelatin/cellulose acetate fibers. The analysis of FTIR spectra showed that the structural changes of gelatin and cellulose acetate occurred after being transformed into gelatin/cellulose acetate nanofibers.

Introduction

Electrospinning is one of the methods used to produce nano to micrometer-sized fibers [1]. By adjusting process parameters, such as voltage, distance between nozzle and collector, nozzle shape, flow rate of the polymer solution, or by changing polymer solution parameters, such as conductivity, viscosity, and surface tension, the fibers morphology and size can be predicted qualitatively [1,2].

Gelatin is a natural polymer derived from collagen composed of peptides and proteins, which is soluble in water [3], similar to collagen, but less expensive [1]. It is generally applied for tissue engineering applications [4-6], wound dressing [7], and drug carrier [8,9]. Although gelatin is widely used for various purposes, it can easily be damaged by water, so that it needs to be blended [5,6,7,10] or cross-linked [11] with other polymers. Cellulose acetate, which is a natural polymer that belongs to the polysaccharide derived from cellulose in many plant cell walls [1], is an alternative for blending.

The solvent selection is important, regarding to the possible applications of fibers in the biomedical field, such as for drug carrier. However, in several studies, the solvents used are toxic, such as 1,1,1,3,3,3-hexafluoro-2-propanol, trifluoroacetic acid, dymethylformamide (DMF), dichloromethane (DCM), dimethylacetamide (DMAc), and chloroform [2,12]. So far, acetic acid, which is less toxic, had been used as a solvent for coaxially electrospun gelatin/cellulose acetate fibers [9,10]. However, it has not been used to produce gelatin/cellulose acetate fibers using simple electrospinning. Here, we report the simply electrospun gelatin/cellulose acetate fibers and their physico-chemical characteristics.

Materials and Methods

Cellulose acetate (average Mn ~50000 and 39.20 – 40.20 wt% of acetyl) was purchased from Sigma-Aldrich. Linear gelatin, made from marrows and bones of cow, was purchased from Bratachem. Glacial acetic acid was used as the solvent. Gelatin and cellulose acetate were dissolved in acetic acid at a concentration of 15 wt%. The mass ratio of gelatin to cellulose acetate was varied at 10:0, 8:2, and 6:4. Each polymer solution was stirred with a magnetic stirrer at 600 rpm for 3 hours at room temperature.

The polymer solution was then transferred into a syringe (Terumo®, capacity 10 mL) with an inner diameter of 15 mm. The syringe has a nozzle with length of 38 mm and diameter of 0.8 mm. The polymer solution was processed by using electrospinning at a voltage of 11 kV, a solution flow rate of 0.1 mL/h, and a nozzle-collector distance of 10 cm. The electrospinning chamber was kept at a relative humidity of 50-55% and a temperature of 30-33°C.

Morphology of gelatin/cellulose acetate fibers was characterized by using a scanning electron microscope (SEM JEOL-JSM-6510LA) with a voltage of 5 kV. Gelatin/cellulose acetate was coated with gold before being characterized. The SEM images of the fibers were analyzed by using software Image MIF v3.0. Meanwhile, functional groups in gelatin/cellulose acetate fibers were analyzed by using a Fourier Transform Infrared (FTIR) spectrometer (Bruker Alpha 1-176-396) with spectral range of 500-4000 cm^{-1} . Moreover, viscosity, surface tension, and conductivity of the polymer solution were measured by using Ostwald Viscometer (Fisher), du Nouy Tensiometer (Fisher), and Conductometer (Mettler Toledo), respectively. The physical properties of the polymer solution were measured at a temperature of 25°C.

Results and Discussion

The SEM images of gelatin/cellulose acetate fibers at 10:0, 8:2, and 6:4 are shown in Figs. 1(a)-1(c). Their diameter distributions are depicted in Figs. 1(d)-1(f), respectively. As given in Table 1, the average fibers diameter increases with the increase of cellulose acetate concentration.

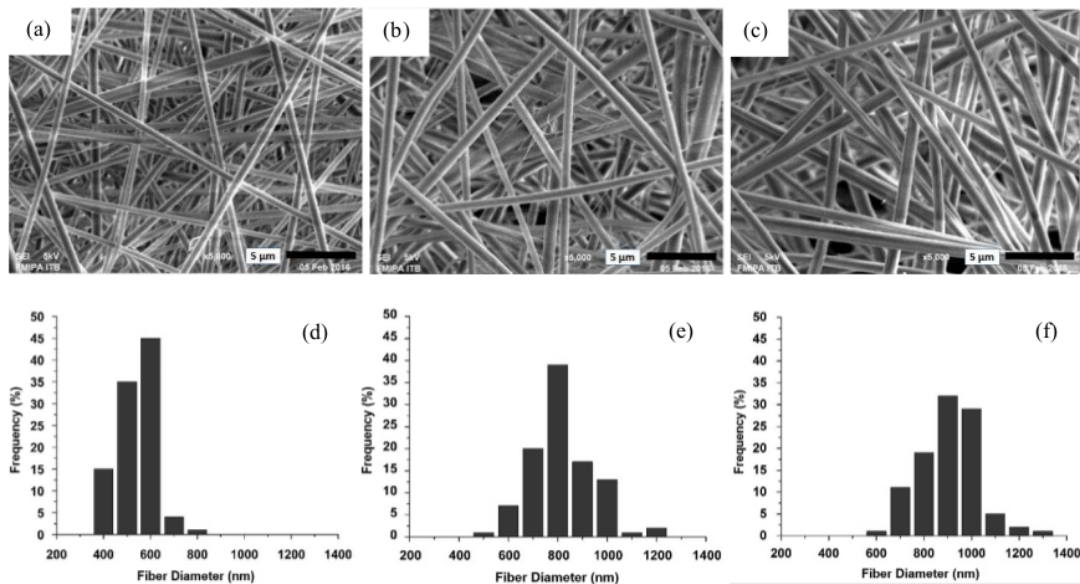


Fig. 1. SEM images (a-c) and diameter distributions (d-f) of gelatin/cellulose acetate fibers from the precursor solutions of 10:0, 8:2, 6:4, respectively.

Table 1 also shows the viscosity, conductivity, and surface tension of the precursor solutions. It can be seen that a higher solution viscosity resulted in a larger size of fibers. Meanwhile, a higher conductivity of solution resulted in a smaller size of fibers. For the solution surface tension, the difference among them was not very far but it shows a tendency that the higher the viscosity of precursor solution, the lower its surface tension.

The addition of cellulose acetate solution into gelatin solution significantly increased the viscosity of gelatin/cellulose acetate solution. It might be caused by the binding between cellulose acetate polymer chains with gelatin polymer chains. The more cellulose acetate added, the more cellulose acetate polymer chains could bind to gelatin polymer chains, causing the increase of gelatin/cellulose acetate solution viscosity. When this solution was being spun, there were more bonds of gelatin/cellulose acetate chains that could hinder the thinning of the fibers being attracted towards the collector. Consequently, the fibers had larger size.

Table 1. Physical properties of gelatin/cellulose acetate fibers and their precursor solutions.

Gelatin/Cellulose Acetate	Average Diameter of Fibers [nm]	Viscosity [cP]	Conductivity [$\mu\text{S}/\text{cm}$]	Surface Tension [dyne/cm]
10:0	534 \pm 25.3	62.924	172.9	41.29
8:2	649 \pm 21.0	176.836	46.2	39.55
6:4	765 \pm 134.3	412.628	33.5	36.45

The precursor solution conductivity of gelatin/cellulose acetate at 10:0 was highest, but then decreased as the amount of gelatin was decreased. It is related to the fact that gelatin is a polyelectrolyte polymer that has a lot of ionized groups, for example amine and carboxyl acids [3,10]. Thus, it can be easily ionized, especially when dissolved in an organic acid solvent. The amine and carboxyl acids group were identified by using FTIR analysis as given later. The less gelatin dissolved in acetic acid, the less ions formed in the solution, resulting in smaller resultant of electrical forces due to the charge interaction. Moreover, the polymer solution was weakly pulled towards the collector, so that the jet was softly stretched and the fibers size became bigger [1].

The increase of viscosity or the reduction of conductivity of the precursor solution increased the average diameter of the gelatin/cellulose acetate fibers. These results are consistent with several other studies [7,8].

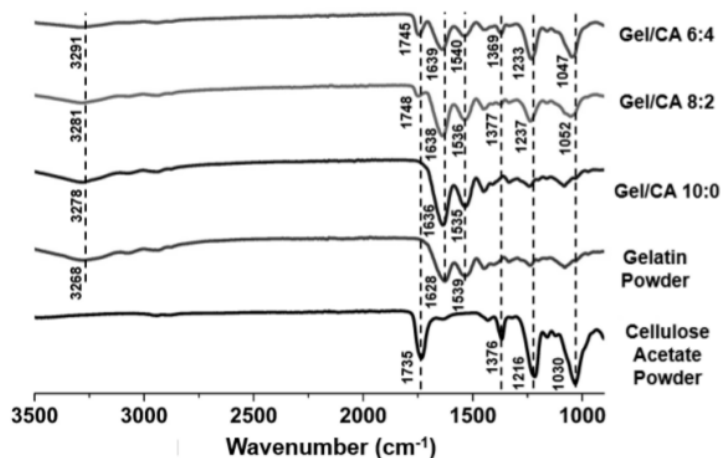


Fig. 2. FTIR transmission spectra of gelatin, cellulose acetate, and gelatin/cellulose acetate fibers.

Several specific peaks of gelatin observed at 3268, 1628, and 1539 cm^{-1} , as shown in Figure 2, represent amide A related to stretching vibration of N-H bond and O-H bond of carboxyl acid, amide I indicated by the stretching vibration of C=O of peptide linkages in the backbone of protein, and amide II related to the coupling of N-H bond and stretching vibration of C-N bond, respectively [6,7,13]. Specific peaks at 1735, 1376, 1216, and 1030 cm^{-1} are identified as the stretching vibration of C=O bond in the ester group of cellulose acetate, the presence of methyl group within acetate group of cellulose acetate, asymmetric stretching of C=O bond within ester group, and the presence of pyranose ring of cellulose acetate, respectively [7,8]. Generally, when compared to the bulk gelatin, specific peaks of the gelatin/cellulose acetate fibers shifted towards higher wavenumbers. It indicates that intermolecular interaction in the gelatin/cellulose acetate fibers is stronger than that of the bulk gelatin due to the presence of cellulose acetate.

Conclusion

Gelatin/cellulose acetate fibers have been produced by using simple electrospinning. By analyzing their SEM images, it has been shown that the average diameters of gelatin/cellulose acetate fibers from the precursor solutions of 10:0, 8:2, and 6:4 were 534, 649, and 765 nm, respectively. The addition of cellulose acetate has increased the viscosity of gelatin/cellulose acetate solution, thus yielding larger fibers diameter. Gelatin mass reduction has caused a decrease of gelatin/cellulose acetate solution conductivity due to the decrease of amine and carboxyl group molecules that can be ionized. Therefore, the increase of viscosity or the reduction of conductivity of the precursor solution has increased the average diameter of the gelatin/cellulose acetate fibers. The obtained FTIR spectra have indicated that there were changes in the structure of gelatin and cellulose acetate after being fibers.

Acknowledgment

This research was financially supported by Directorate of Research and Community Engagement of Ministry of Research, Technology and Higher Education, Indonesia under the University's Excellent Research (PUPT) Grant in the fiscal years of 2015-2016.

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