The Relation of Rheological Properties with Electrospinnability of Gelatin/Sodium Alginate Solutions

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Abstract

The present study aims to reveal the viscosity dependence of fiber diameter and morphology. For this purpose, the rheological behavior of gelatin/sodium alginate solutions, prepared at different conditions (i.e., polymer concentration, solvent composition, and blending ratio), was analyzed by using a rheometer. Meanwhile, the fiber diameter and morphology of electrospun gelatin/sodium alginate nanofibers obtained at different conditions were investigated by scanning electron microscope. Employing both experimental data, the relation between viscosity and spinnability was evaluated. Results showed that viscosity increased with the concentration of gelatin, sodium alginate, and acetic acid. Since an increase in viscosity is generally associated with fibers having larger diameter and better morphology, it was of great importance to determine the impact of solution parameters on viscosity. This study provides an overview on the fabrication of gelatin/sodium alginate nanofibers which may have potential to be used in the field of tissue engineering.

1. Introduction

Electrospinning is a simple and cost-effective technique through which fibrous mats with diameters ranging from several microns down to a few nanometers can be fabricated from both synthetic and natural polymers [1–3]. In the last decade, this technique has gained much attention for biomedical applications, including tissue engineering scaffolds, wound dressing pads, and drug delivery platforms [4–7].

The morphology and diameter of electrospun fibrous mats depend on many parameters [2, 4–7]. Since the characteristics of these mats are associated with their morphology and fiber diameter [3, 4, 7], it is very crucial to assess the influence of these parameters. Among them, viscosity is a key factor in the electrospinning process, since it determines whether the jet breaks up into droplets, beads, or fibers. On the other hand, higher viscosity resists on the extension of the jet, increasing fiber diameter.

The synthesis of natural polymer-based nanofibers is of interest because of their many outstanding properties, including biological origin, biocompatibility, biodegradability, hydrophilicity, commercial availability, renewability, and cost efficiency [8–10]. To date, previous researchers have shown the potential use of gelatin/sodium alginate-based materials for biomedical applications, including drug delivery [11, 12], wound healing [13–15], and tissue engineering [16, 17]. To the best of our knowledge, no systematic study has been reported to evaluate the relationships between the rheology and the electrospinnability of gelatin/sodium alginate nanofibers. Therefore, the main objective of the present work was to determine the influence of gelatin concentration, sodium alginate concentration, solvent composition, and blending ratio on rheological behaviors of gelatin/sodium alginate solutions and how this relationship affects the morphology and diameter of electrospun fibrous mats.

2. Experimental Procedure

Gelatin (type A, from porcine skin) and sodium alginate (alginic acid sodium salt from brown algae) were obtained from Sigma–Aldrich Chemicals. Glacial acetic acid was purchased from Merck. All chemicals were used as provided without further purification.

Gelatin solutions with concentrations of 10%–20% (w/v) were first prepared by dissolving in 40%–80% (v/v) acetic acid aqueous solutions at room temperature for 2 h. Meanwhile, sodium alginate was dissolved in deionized water at room temperature for 24 h to obtain the alginate solutions with concentrations of 0–2 wt.%. Afterward, gelatin and alginate solutions were mixed at different volumetric ratios.

The blend solutions, which were transferred into a 5 mL syringe, were delivered via a syringe pump to maintain a steady flow of the solution at 3 mL/h. Electrospinning was conducted under a constant applied voltage of 20 kV. Randomly oriented electrospun fibers were collected on a grounded plate wrapped with aluminum foil, which was placed at a distance of 10 cm from the syringe tip. All electrospinning experiments were performed at ambient conditions.
The rheological measurements used to characterize the spinning solutions were performed using a rheometer (RM180 Rheomat, Rheometric Scientific). Steady shear measurements were carried out at 25°C in the range of 15–1200 s⁻¹.

The surface topography and fiber diameter of the as-spun fibrous mats were determined with the aid of a scanning electron microscope (SEM, JSM-5410, Jeol). Prior to imaging, a small section of the samples cut from the fibrous mats was sputter coated with platinum by using a sputter coater (SC7620, Quorum Technologies Ltd) for 120 s. For each experiment, the average fiber diameter and its standard deviation were analyzed by the help of an image visualization software (Image-J, National Institute of Health) from about 50 measurements of the random fibers.

3. Results and Discussion

3.1 Effect of gelatin concentration

To investigate the dependence of rheological and electrospinning properties on gelatin concentration, three different blend solutions with varying gelatin concentrations (10, 15, and 20% w/v) were employed. Figure 1 depicts the dependence of viscosity on gelatin concentration. The solutions with higher gelatin concentrations showed higher viscosities, as might be expected for common polymeric solutions.

![Figure 1. The dependence of viscosity on gelatin concentration.](image1)

As observed from Figure 1, the viscosity of the solution with a gelation concentration of 10% w/v was almost independent of shear rate and the solution behaved as a Newtonian fluid. However, the solution exhibited a transition from Newtonian fluid to non-Newtonian fluid as the gelatin concentration increased. As seen from Figure 2, the morphology of the electrospun nanofibers changed from beads-on-fiber to bead-free fibers as the shear thinning behavior of the solutions increased. Meanwhile, the average diameter of the electrospun nanofibers increased from 106 to 124 nm with the increase in gelatin concentration from 10% to 20% w/v. This matched with the rheological measurements showing the increase in viscosity with the increase in gelatin concentration.

![Figure 2. Representative SEM images of gelatin/sodium alginate nanofibers with varying gelatin concentrations.](image2)

3.2 Effect of sodium alginate concentration

To evaluate the dependence of rheological and electrospinning properties on sodium alginate concentration, three different blend solutions with varying sodium alginate concentrations (0, 1, and 2 wt.%) were used. Figure 3 displays the dependence of viscosity on sodium alginate concentration. It is obvious that the solution with no sodium alginate (0 wt.%) behaved as a Newtonian fluid, while the solutions with sodium alginate (1 and 2 wt.%) showed a non-Newtonian behavior. Additionally, the shear thinning behavior of solutions increased with the sodium alginate concentration, which led to a change from beads-on-fiber to bead-free fibers in the morphology of the electrospun nanofibers (Figure 4). This observation was similar to the one expressed above.

On the other hand, it was determined that the viscosity of solutions with the sodium alginate concentrations of 0 and 2 wt.% were quite similar at higher shear rates (800–1000 s⁻¹, deformation rates felt in the Taylor cone and jet [18]). This was likely the reason for average diameter of the electrospun nanofibers fabricated from these solutions were almost same. Meanwhile, fibers with thicker diameters were obtained when the sodium alginate concentration was 1 wt.%. This may be because the viscosity of this solution was higher at the higher shear rates.
3.3 Effect of acetic acid content in the solvent

To ascertain the effect of solvent composition on solution rheology and spinnability, solvent mixtures of acetic acid and water with different acetic acid contents were utilized to dissolve gelatin. Figure 5 shows the effect of acetic acid content on viscosity of the gelatin/sodium alginate solutions. The solutions with higher acetic acid contents showed higher viscosities, which was likely the reason for however, the solution exhibited a transition from Newtonian fluid to non-Newtonian fluid as the acetic acid content increased. As seen from Figure 6, the morphology of the electrospun nanofibers changed from beads-on-fiber to bead-free fibers as the shear thinning behavior of the solutions increased, matching with observations above.

On the other hand, the viscosity of the solution with the acetic acid content of 40 vol.% was almost independent of shear rate and the solution behaved as a Newtonian fluid.

3.4 Effect of blending ratio

To determine the effect of blending ratio on solution rheology and spinnability, three different blend solutions with varying gelatin/sodium alginate ratios (90/10, 80/20, and 70/30, v/v) were employed. Figure 7 shows the effect of blending ratio on viscosity of the gelatin/sodium alginate solutions. The solutions with higher gelatin contents showed higher viscosities, which was likely the reason for
increasing in fiber diameter from 136 to 115 nm (Figure 8(c) and (a), respectively).

Figure 7. The dependence of viscosity on blending ratio.

On the other hand, the viscosity of each solution was almost independent of shear rate and thus, the solutions exhibited a Newtonian fluid behavior. This was why the morphology of the electrospun nanofibers were beads-on-fiber.

Figure 8. Representative SEM images of gelatin/sodium alginate nanofibers with varying blend ratios.

4. Conclusion

This study aimed to evaluate the relationship between the rheological behavior and the electrospinnability of gelatin/sodium alginate solutions prepared at different conditions (i.e., polymer concentration, solvent composition, and blending ratio). Results showed that viscosity increased with the concentration of gelatin, sodium alginate, and acetic acid. Since the higher the viscosity the larger the fiber diameter is, thicker fibers were obtained when the gelatin concentration, sodium alginate concentration, and acetic acid content were higher. Additionally, Newtonian behavior was associated with beads-on-fiber morphology, whereas solutions behaving as non-Newtonian fluids led to bead-free fibers. Moreover, the surface morphology improved as the shear thickening increased. Consequently, this study provides an insight for future researchers who aim to fabricate fibrous mats through electrospinning technique.

References