EMULSION ELECTROSPINNING OF POLYVINYLPIRROLIDONE (PVP)/PARACETAMOL NANOFIBERS

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Abstract: This study aimed to achieve Polyvinylpyrrolidone (PVP) nanofiber production including paracetamol (PCT) by oil-in-water emulsion electrospinning. At first, emulsions were prepared at 14 wt % PVP with various PCT concentrations (0, 0.1, 0.3, 0.5, 0.7, 0.9 wt %). Then, solution properties such as viscosity, conductivity, and surface tension were determined. The production of nanofiber samples was carried out by emulsion electrospinning under the optimum process parameters (voltage, distance between electrodes, feed rate, and atmospheric conditions). Finally, the morphological and structural characterization of the nanofiber surface was carried out with SEM and FT-IR. According to the results of emulsion properties, although the change is not remarkable, it tends to increase the viscosity with an increase in PCT concentration. On the other hand, it was observed that surface tension did not change significantly with PCT concentration increase and while the conductivity of emulsions decreased slightly. When the fibre structure was investigated, average fibre diameter and fibre diameter uniformity were not affected prominently by PCT concentration. From the SEM images, it is possible to say that generally fine, uniform and bead-free drug-loaded nanofibers were obtained. The finest (326 nm) and most uniform (1.03) nanofibers were achieved from the sample N4 which included 0.5 wt % PCT. Also, the FT-IR results verified that PVP and PCT exist in the nanofiber structure.

Keywords: emulsion electrospinning, nanofiber, paracetamol, Polyvinylpyrrolidone

1. INTRODUCTION

In this study, it was aimed at achieving Polyvinylpyrrolidone (PVP) nanofiber production including paracetamol (PCT) by oil-in-water emulsion electrospinning and characterization in terms of fibre morphology by Scanning Electron Microscope (SEM) and chemically exhibition of PCT presence in the structure of nanofibers by FT-IR analysis.

It is known from the literature that nanofibers can be used in several fields, such as medical applications, drug delivery systems, energy storage, battery separators, aerospace, information technology, etc. [1]. Recently, there have been lots of possible biomedical uses for nanofibers, owing to their unequalled mechanical strength, elasticity, very small fibre diameter, small controllable pore size, high porosity, very high specific surface area (m²/g), high load capacity, and ease of fabrication [2,3].

Emulsion electrospinning is an efficient and new method for the production of nanofibers. From immiscible liquids such as essential oils and hydrophobic drugs and

proteins can be produced as nanofibers by this new approach. The preparation of homogenous and stable emulsion solutions to produce nanofibers is much easier with emulsion electrospinning. Another advantage of this method is that there is no need for an extra apparatus compared to the classical electrospinning system [4].

PVP is a highly hydrophilic polymer and is used because of its remarkable properties, such as high surface activity, good biocompatibility, low toxicity, and strong adsorption capacity. PVP based nanofibers have a wide variety of applications in biomedical, pharmacy, food and cosmetics [5].

PCT is a non-inflammatory [6], antipyretic and analgesic agent which is used to relieve pain in the body and to reduce body temperature [7]. Recently, there has been research about electro spun nanofibers, including PCT. Yang et al. prepared PCT loaded poly (vinyl alcohol) (PVA) nanofibers by electrospinning [8]. Yıldız and Uyar successfully obtained free-standing, easy to handle nanofibrous films of paracetamol/HPCD-IC and paracetamol/SBE--CD-IC by electrospinning [9]. Illangakoon et al., successfully produced fast-dissolving drug delivery systems for the simultaneous release of paracetamol and caffeine. This work was achieved by processing them into electro spun fibres using PVP as the filament forming agent [7].

It is thought that the results obtained from this study will contribute to the literature on medical textile materials.

2. MATERIALS AND METHODS

In this study, PVP (360.000 g/mol) (Sigma–Aldrich (St. Louis, MO, USA) was used as a polymer, PCT was used as a drug (Atabay Pharmaceuticals and Fine Chemicals Inc.), dimethylformamide (DMF) (Sigma–Aldrich (St. Louis, MO, USA) and distilled water were used as solvents. In addition, Cremophor RH 40 was used as a surfactant and was supplied by Ersa Chemistry (Izmir, Turkey).

PVP concentration and surfactant concentration were kept constant at 14 wt % and 1 wt %, respectively. PCT was added at different concentrations, such as 0.1, 0.3, 0.5, 0.7, and 0.9 wt%.

According to the experimental studies, initially, polymer solutions were prepared as an emulsion with PVP and PCT. The emulsion was stirred at room temperature for about 20 hours. After emulsion preparation, the emulsion properties were determined. The conductivity, viscosity, and surface tension (by the Du Noüy ring) of emulsions were measured.

Then, the nanofibers were produced by an emulsion electrospinning approach. The process parameters of the electrospinning given in table 1, such as applied voltage, distance between needle and collector, solution flow rate etc., were optimized. All process parameters were kept constant as in table 1 during the spinning process. Also, the spinning process was achieved under the same ambient conditions for all samples. The nanofibers were produced for 20 minutes and collected on an aluminium sheet.

Table 1. Process parameters of the electrospinning

Voltage (kV)	Distance between electrodes (cm)	Feed rate (ml/h)	Humidity (%)	Temperature (°C)
21	19.5	0.3	28	25

Finally, the nanofiber morphology, such as fibre diameter, diameter uniformity, and web surface structure, was determined and analysed with SEM. The fibre diameters

were calculated by image J analysing software. The SPSS analysis program was used to statistically analyse the findings about morphological parameters such as fibre diameter histogram. FT-IR was performed to determine the presence of PVP and PCT in the nanofiber structure chemically.

3. RESULTS AND DISCUSSIONS

In this study, emulsion properties such as viscosity, conductivity, and surface tension were measured and the effects of PCT concentration on the solution properties were analysed. From figure 1, it was determined that there was no significant change in surface tension, viscosity, and conductivity with different PCT concentrations.

According to the results of emulsion properties, although the change is not significant, it tends to increase the viscosity with an increase in PCT concentration except N6.

On the other hand, it was observed that surface tension tends to decrease as the PCT concentration increases, except for the N4 sample, but the changes are not significant. The conductivity of emulsions decreases with PCT increase except N2. It is possible to say that emulsion conductivity increases with the first addition of PCT to the PVP emulsion and then decreases as the PCT concentration increases.

Figure 1. Results of solution properties (surface tension, viscosity and conductivity) of PVP with various PCT concentrations.

SEM images and fibre diameter histograms of PVP nanofiber samples (1.000x, 10.000x) produced with various PCT concentrations are given in figure 2.

(N1)

(N2)

(N3)

(N4)

(N5)

(N6)

Figure 2. SEM images and fibre diameter histograms of PVP nanofiber samples produced with various PCT concentrations (1.000x, 10.000x)

When figure 2 was analysed, it was seen that uniform and smooth nanofibers, good web quality without beads, and unimodal fibre diameter histograms were obtained. There was no bead formation in any fibre structure and the PCT concentration did not affect the bead formation.

The effect of PCT concentration on the average fibre diameter and fibre diameter

uniformity coefficient was shown in figure 3.

Figure 3. The effect of PCT concentration on the average fibre diameter and fibre diameter uniformity coefficient of PVP nanofibers

According to Figure 3, there isn't any specific variation in the fibre diameter and diameter uniformity coefficient with change in PCT concentration. The smallest average fibre diameter is 326 nm (N4) and the highest one is 385 nm (N6) for all fibre structures among all PCT concentrations. Fibre diameter uniformity coefficient values are very close to 1 for all samples. The smoothest histogram was again seen in the N4 sample.

Lastly, the presence of PVP and PCT were confirmed in the nanofiber structure via FT-IR analysis (Figure 4). According to FT-IR spectra of the PVP, there is a peak at 2949 cm^{-1} . This peak appeared at 2949 cm^{-1} in N1, and 2953 cm^{-1} in N4. There is another sharp peak at 1652 cm^{-1} due to the C=O in the spectra of the PVP polymer. This peak appeared at 1657 cm^{-1} and 1655 cm^{-1} in the spectra of N1 and N4 respectively. Due to the C–N stretch from PVP, there is a peak at 1284 cm^{-1} . This peak appeared at 1288 cm^{-1} in both N1 and N4. On the other hand, pure PCT has a peak at 1561 cm^{-1} . This peak appeared at 1559 cm^{-1} in an N4 sample that included 0.5 wt %.

Figure 4. FT-IR spectra of PVP, PCT and PCT loaded PVP nanofiber samples

4. CONCLUSION

The production and characterization of PVP nanofibers with various concentrations of PCT were achieved via an emulsion electrospinning approach. According to the results, it was observed that quite uniform, smooth and fine, drug-loaded emulsion electro spun nanofibers were produced. In addition, good web quality without beads and a unimodal histogram curve of fibre diameter were determined. It is thought that this nanofibrous medical textile surface obtained has the application potential for controlled drug delivery applications.

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