Nanofiber production from rose water and mate plant extract solutions using environmentally friendly electrospinning

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ABSTRACT – REZUMAT

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In this study, it was aimed to produce nanofibers from polyvinyl alcohol (PVA) polymer using natural solvents such as rose water, rose extract and mate plant extract by the environmentally friendly electrospinning or green electrospinning technique in other words and it was also aimed to investigate the morphological properties of produced nanofibers. For this purpose, nanofibers having 7 different morphologies were produced from 4% PVA aqueous solutions. The morphologies of the produced nanofibers were analysed by scanning electron microscopy (SEM). As a result of these analyses, it was observed that uniform nanofiber morphology was formed in nanofiber productions made with distilled water, while in the others, dense bead structure was formed at low voltages and nanofiber morphology with reduced bead amount at high voltages was observed. In all electrospinning experiments, it was observed that the nanofibers were randomly collected on the collector plate. It was observed that the nanofibers obtained from the solutions prepared from solvents other than distilled water, especially the one from the mate plant extract, had a low bead structure and a smooth morphology. These results showed that environmentally friendly nanofibers can be produced from natural solvents by electrospinning technique and their morphological properties can be improved via modification in process conditions. It is thought that the nanofibers produced within the scope of the study are candidate materials especially for health, hygiene and food applications since they are produced in pure properties without using any other additives.

Keywords: *green electrospinning, nanofibers, rose water, mate, PVA*

Producerea de nanofibre din soluții de apă de trandafiri și extract de plantă mate utilizând electrofilarea ecologică

În acest studiu, s-a urmărit producerea de nanofibre din polimer de alcool polivinilic (PVA) folosind solvenți naturali, cum ar fi apa de trandafiri, extractul de trandafiri și extractul de plantă mate, prin tehnica de electrofilare ecologică sau electrofilare „verde" și, de asemenea, s-a urmărit investigarea proprietăților morfologice ale nanofibrelor produse. În acest scop, din soluții apoase 4% PVA au fost produse nanofibre având 7 morfologii diferite. Morfologiile nanofibrelor produse au fost analizate prin microscopie electronică cu scanare (SEM). În urma acestor analize, s-a observat că s-a format o morfologie uniformă în producțiile de nanofibre realizate cu apă distilată, în timp ce în celelalte s-a format o structură densă a perlelor la tensiuni joase și s-a observat morfologia nanofibrelor cu cantitate redusă de perle la tensiuni înalte. În toate experimentele de electrofilare, s-a observat că nanofibrele au fost colectate aleatoriu pe placa colectoare. S-a observat că nanofibrele obținute din soluțiile preparate din alți solvenți decât apa distilată, în special cea din extractul de plantă mate, aveau o cantitate redusă de perle și o morfologie netedă. Aceste rezultate au arătat că nanofibrele ecologice pot fi produse din solvenți naturali prin tehnica de electrofilare și proprietățile lor morfologice pot fi îmbunătățite prin modificarea condițiilor de proces. Se crede că nanofibrele produse în cadrul studiului sunt materiale destitate în special pentru domeniile sănătate, igienă și aplicații alimentare, deoarece au proprietăți pure, fără a utiliza niciun alt aditiv.

Cuvinte-cheie*: electrofilare ecologică, nanofibre, apa de trandafiri, mate, PVA*

INTRODUCTION

Nanofibers are engineering materials with diameters below 1μm, a large surface area and a high number of interconnected pores [1]. These superior properties have made nanofibers an important candidate for a wide variety of applications such as medical textiles, filtration, isolation, sensing, aerospace, tissue engineering and energy applications [2, 3]. The electrospinning technique has become the most preferred method in the production of nanofibers in the last 20 years, but toxic solvents harmful to nature and human health have been used in many studies. In this context, the nature and human-friendly electrospinning method has gained importance recently, both for political and economic reasons and due to the demands of customers in this direction [4]. Deionized water and acetone have become widely used solvents in nanofiber production with this method which is also called green electrospinning.

Polyvinyl alcohol (PVA) is one of the most commonly used polymers in this method. Researchers produced nanofibers by electrospinning method using PVA polymer and deionized water and reported that the nanofibers obtained as a result of the analyses were hygienic and environmentally friendly [5, 6]. In another study, researchers produced polyelectrolyte nanofibers with the help of water from a polymer solution of PVA, polyacrylic acid (PAA) and polyethylene imine (PEI) using the electrospinning method, and it was reported that the nanofibers have shown a good antimicrobial activity and an acceptable degree of strength according to the infrared (IR), scanning electron microscope (SEM) [7]. Bosworth and Downes (2012) developed environmentally friendly nanofibers with ideal morphological properties using the electrospinning method with the help of a sustainable acetone solvent [8].

In recent years, apart from PVA polymer, green electrospinning has been realized by using many different polymers such as polyamide 11 (PA 11) [9], chitosan [10], polycaprolactone (PCL)/collagen [11], polyvinylidene fluoride (PVDF)/ polyvinylpyrrolidone (PVP) [12] and polyethylene oxide (PEO) [13]. Apart from electrospinning, there are also studies in which environmentally friendly nanofiber production is carried out by the solution-blowing method, but these studies have not become widespread yet [14, 15].

The production of nanofibers with an environmentally friendly electrospinning technique is relatively common and promising in terms of many functional applications. Some of these applications are health, biomedical and drug distribution [16]. In the field of drug delivery, researchers have developed environmentally and human-friendly nanofiber structures that can distribute natural active substances to provide antimicrobial activity [17] and antioxidant activity [18] with the help of the green electrospinning technique. In another study, researchers developed nanofiber scaffolds using collagen/hydroxyapatite materials with the help of the green electrospinning technique. Another application area is packaging [20, 21]. In a study using pure water as a solvent, the researchers developed nanofibers for the active packaging area with the help of electrospinning [22]. Another application area is filtration [23]. In the article published in the journal Nature Scientific Reports, researchers have produced silk protein nanofibers for highly efficient, environmentally friendly, semi-transparent multifunctional air filters using the electrospinning technique [24].

In this study, the production and characterization of environmentally friendly nanofibers from PVA polymer were carried out using solvents obtained entirely from local sources in Turkey, such as rose water, rose extract, mate plant extract and pure water, which are described as environmentally friendly and green. To the best of our knowledge, no other study was found in which mate plant extract and rose extract extracted from the rose pulp harvested from the field were used as a solvent in the production of nanofibers.

EXPERIMENTAL WORK

Materials

In this study, PVA (100.000 g/mol) obtained from Sigma Aldrich company was used as the polymer. Four different solutions were prepared. In the first, commercial rose water, which was completely supplied from Turkey's domestic resources as a solvent, was obtained from Isparta Gülbirlik Cooperative. The first solution was prepared by adding 96 ml of rose water to 4 g of PVA polymer (4% wt) using this rosewater. For comparison, rose extract (pure rose water) was obtained by collecting roses directly from the rose fields in the Senir district of Isparta during the harvest season and keeping them in pure water at approximately 90°C and used within the scope of the study. For this second solution, 10 g of fresh rose leaves were extracted by adding 100 ml of hot water, and then a solution was prepared by adding 96 ml of pre-prepared rose extract to 4 g of PVA polymer (4% wt). For the third solution, the Mate plant, originating from South America, was obtained from a herbalist in Isparta and the extract has been prepared. Mate plant is an antioxidant plant containing 24 vitamins and minerals [25]. For this study, the extract was prepared by adding 10 g of mate plant leaves to 100 ml of hot water. The third solution was prepared by adding 96 ml of pre-prepared mate plant extract to 4 g of PVA polymer (4% wt). Finally, for comparison purposes, the fourth solution was prepared by adding 96 ml of distilled water to 4 g of PVA polymer (4% wt). The properties of the prepared solutions are given in table 1.

Table 1

SOLUTION PROPERTIES USED IN NANOFIBER PRODUCTION

Solution acronym	Polymer and concentration (%)	Solvent and concentration (%)
RWPVA	PVA (4%)	Rose water (96%)
REPVA	PVA (4%)	Rose extract (96%)
MTPVA	PVA (4%)	Mate extract (96%)
DWPVA	PVA (4%)	Distilled water (96%)

Electrospinning

In this study, environmentally friendly nanofiber production was carried out using the electrospinning technique under variable process conditions. Electrospinning is the most widely used technique in the production of nanofibers today. In this technique, production is carried out with a simple mechanism that can be installed at an affordable cost. The electrospinning apparatus consists of an electronically controlled syringe pump, a syringe, a high-voltage power supply and a collector plate. As can be understood from the configuration of the device, principally the polymer fed from the syringe is subjected to an electrostatic force at a certain voltage (kV) and collected on the collector plate in the form of nanofibers [26]. In this study, an Electrospinning device belonging

Table 2

PROCESS PARAMETERS APPLIED IN ELECTROSPINNING			
Voltage (kV)	**Collector-nozzle distance (cm)**	**Polymer feeding rate (ml/h)**	**Solution**
13	6	0.35	DWPVA
13	6	0.45	RWPVA
21	12	0.45	RWPVA
9	6	0.35	REPVA
13	8	0.35	REPVA
19	8	0.35	MTPVA
23	10	0.35	MTPVA

to Inovenso Ltd & Inc. Company originating from Turkey was used. During the operation, the ambient temperature was between 27–32 ºC and the relative humidity was between 35% and 45%. The process parameters applied for electroproduction are given in table 2.

Scanning Electron Microscope (SEM) analysis

Alignment and morphological properties of the obtained nanofibers were characterized using an SEM instrument (LEO 440, Technical Sales Solutions, Beaverton, OR, ABD).

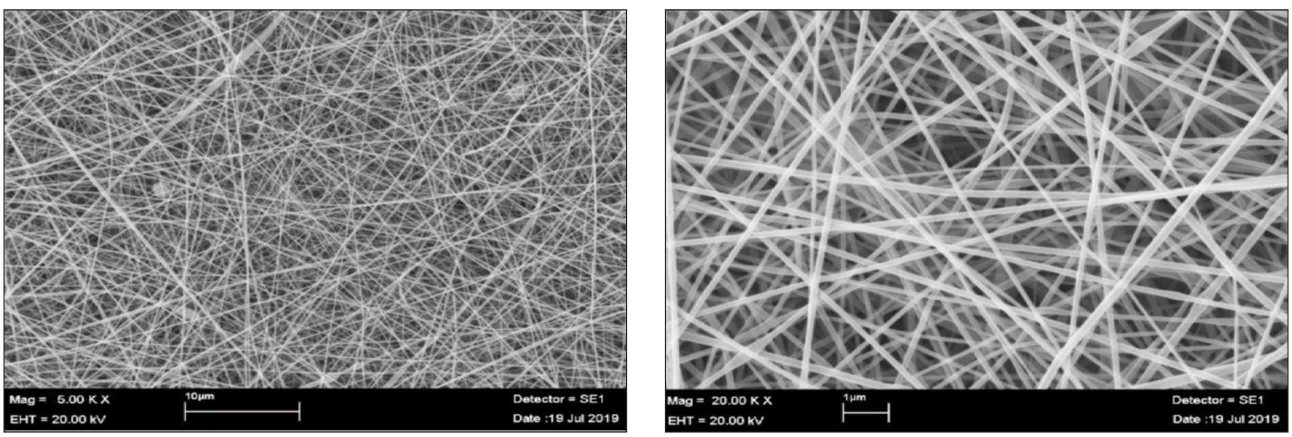
RESULTS AND DISCUSSIONS

In this study, nanofibers were produced from DWPVA, RWPVA, REPVA, and MTPVA solutions, respectively, by the electrospinning method and the morphologies of the produced nanofibers were characterized using SEM.

In figure 1, SEM images of nanofibers produced from the DWPVA solution obtained by dissolving PVA in pure water at 13 kV voltage, 6 cm collector-nozzle distance and 0.35 ml/h process conditions are given. It was observed that the produced nanofibers were randomly collected on the collector and the fibre distribution was randomly oriented. Also, no bead formation was observed.

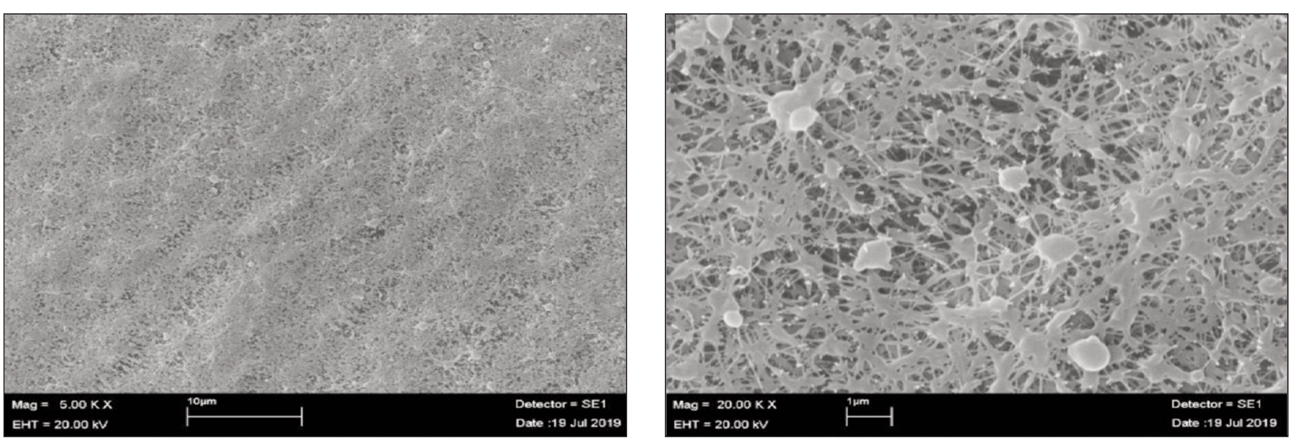
In figure 2, SEM images of nanofibers produced from the RWPVA solution in process conditions of 13 kV voltage, 6 cm collector-nozzle distance and 0.45 ml/h polymer feeding rate were given. When the images were examined, it was observed that although fibre formation was observed, a high number of beads occurred.

In figure 3, SEM images of nanofibers produced from the RWPVA solution in process conditions of 21 kV



a *b*

Fig. 1. SEM images of nanofibers produced from DWPVA solution: *a* – 5 kX; *b* – 20 kX



a *b*

Fig. 2. SEM images of nanofibers produced from RWPVA solution: *a* – 5 kX; *b* – 20 kX

voltage, 12 cm collector-nozzle distance and 0.45 ml/h polymer feeding rate were given. When the images were examined, it was observed that beads also occurred under these process conditions, but the fibre formation was relatively more apparent than in the previous one. This partial improvement in fibre structure is thought to be a result of operating under higher voltage conditions.

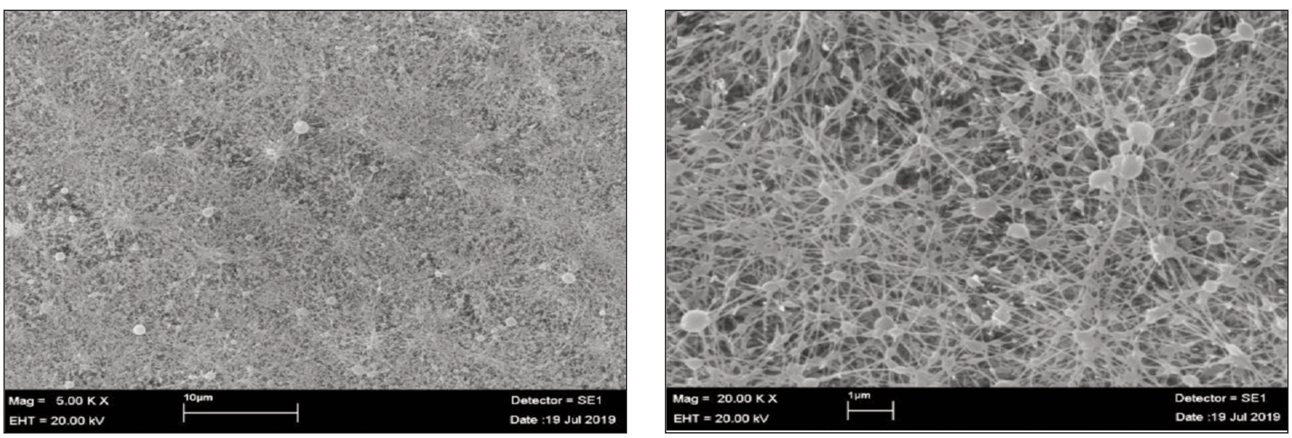
In figure 4, SEM images of nanofibers produced from the REPVA solution in process conditions of 9 kV voltage, 6 cm collector-nozzle distance and 0.35 ml/h polymer feeding rate were given. When the images were examined, it was observed that although fibre formation was observed, intense bead formation also occurred.

In figure 5, SEM images of nanofibers produced from the REPVA solution in process conditions of 13 kV voltage, 8 cm collector-nozzle distance and 0.35 ml/h polymer feeding rate were given. When the images were examined, it was seen that the bead formation decreased slightly with the increase in voltage, and the fibrous structure became more noticeable. The results obtained so far are in accordance with studies by different researchers in the literature and especially confirm the voltage-bead formation relationship [27, 28].

In figure 6, SEM images of nanofibers produced from the MTPVA solution in process conditions of 19 kV voltage, 8 cm collector-nozzle distance and 0.35 ml/h polymer feeding rate were given. When the images were examined, bead formation was observed in the nanofibers, but it was also seen that the fibre structure is evident.

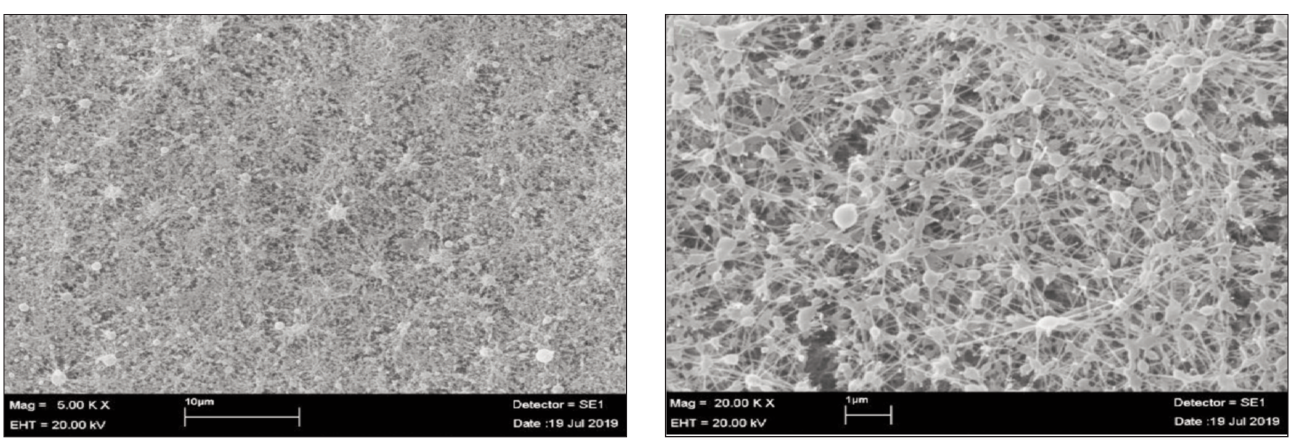
In figure 7, SEM images of nanofibers produced from the MTPVA solution in process conditions of 23 kV voltage, 10 cm collector-nozzle distance and 0.35 ml/h polymer feeding rate were given. When the images were examined, it was seen that the bead formation decreased significantly with the increase of the voltage applied to the electrospinning area and the nanofibers with regular morphology were randomly collected on the collector plate. The results obtained in this part of the study are compatible with the results of the nanofibers produced using the RWPVA solution and the literature information in this field.

In other relevant literature, there are also studies in which structures such as carbon nanotubes are added to the polymer solution, unlike increasing the voltage applied to the electrospinning area to reduce bead formation and enhance the nanofiber structures [29, 30]. Future studies of the current research, it is



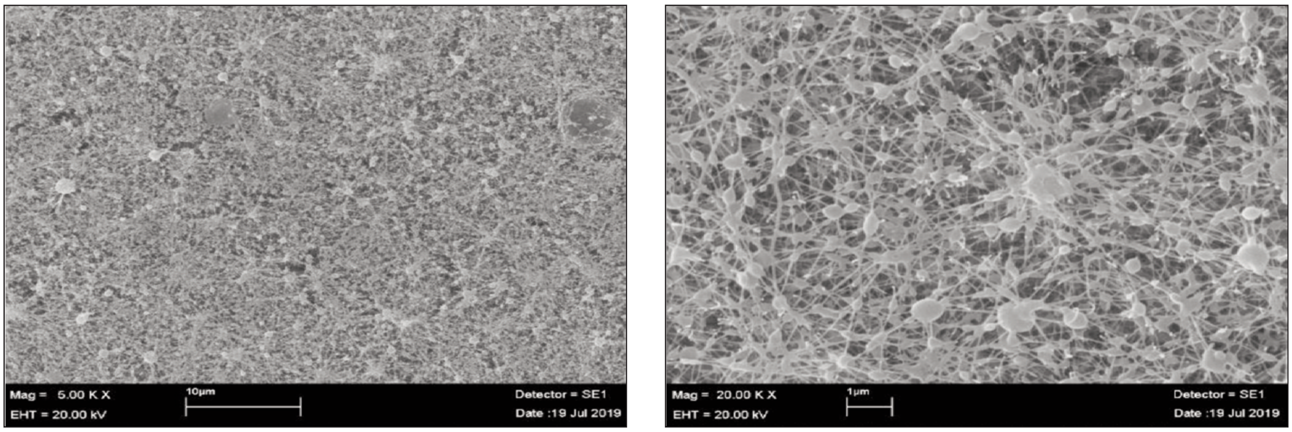
a *b*

Fig. 3. SEM images of nanofibers produced from RWPVA solution: *a* – 5 kX; *b* – 20 kX



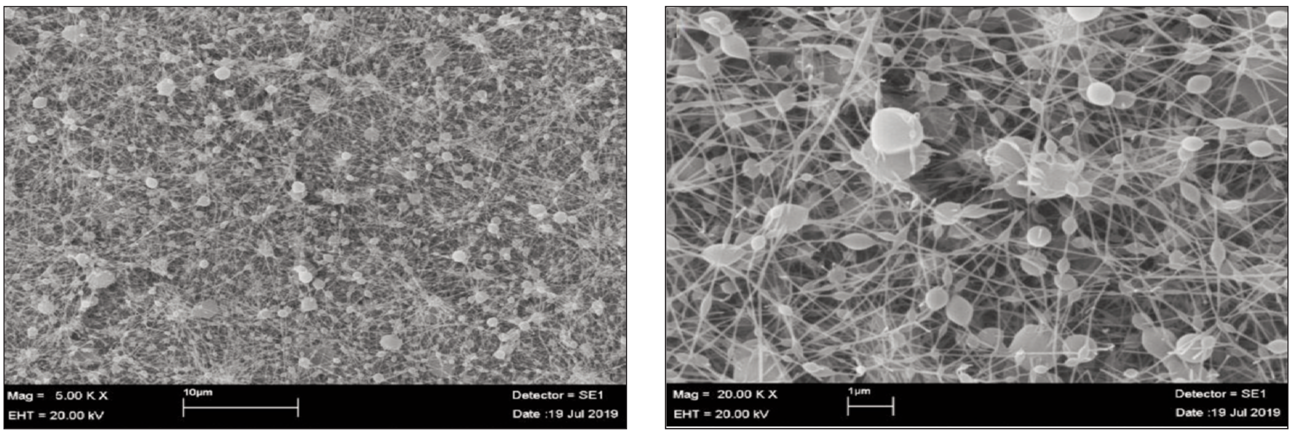
a *b*

Fig. 4. SEM images of nanofibers produced from REPVA solution: *a* – 5 kX; *b* – 20 kX



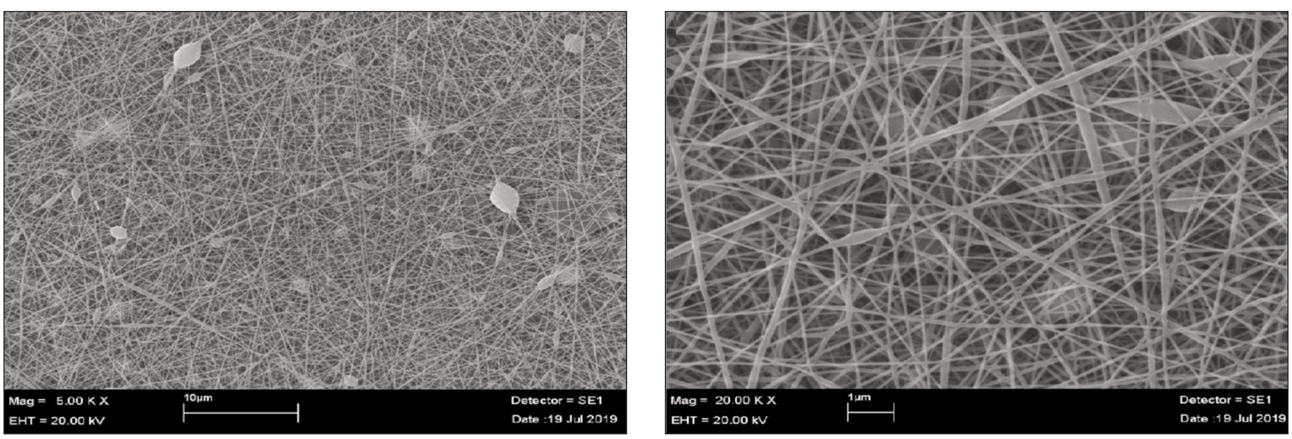
a *b*

Fig. 5. SEM images of nanofibers produced from REPVA solution: *a* – 5 kX; *b* – 20 kX



a *b*

Fig. 6. SEM images of nanofibers produced from MTPVA solution: *a* – 5 kX; *b* – 20 kX



a *b*

Fig. 7. SEM images of nanofibers produced from MTPVA solution: *a* – 5 kX; *b* – 20 kX

aimed to add nature-friendly structures/particles to the solution to diversify and improve the morphology of nanofibers to be produced.

CONCLUSIONS

In this study, environmentally friendly nanofiber production was carried out under varying process conditions (voltage, collector-nozzle distance, feed rate) with the help of the electrospinning technique using commercially available rose water, harvested rose extract and mate plant extract as solvents. To be a reference product, PVA polymer was first dissolved in pure water, then nanofiber production was carried out by electrospinning, and as a result of SEM analysis, it was observed that nanofibers had uniform morphology and fibre distribution. In nanofiber production studies carried out with rose water under 2 different process conditions, a partial decrease in the number of beads that formed in the fibres was observed with the increase of the voltage applied to the electrospinning area. In nanofiber production studies carried out with rose extract under 2 different process conditions, it was observed that pilling occurred in high amounts, especially in low-voltage production and fibre formation came to the forefront with increasing voltage. In nanofiber production studies carried out under 2 different process conditions in which Mate plant extract was used as a solvent, beads formed at low voltage as in previous studies, but a critical decrease was observed in the number of beads when the voltage was increased, and the fibre morphology was found to be almost as uniform as the morphology of the nanofibers obtained in the reference production carried out with distilled water. These results show that especially the applied voltage has critical importance considering process conditions. Environmentally friendly nanofibers were successfully produced from all solvents used in the study. These nanofibers have the potential to be an ideal material for areas such as health, hygiene and food, where naturalness, biocompatibility and sustainability are important and necessary. Since nanofibers produced using mate plant extract have a more uniform morphology, they may have the potential to be used in applications such as drug release, tissue engineering and wound healing where fibre orientation is important.

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