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SARI KANTARON YAĞI KATKILI PVA NANOLİFLERİN EMÜLSİYON ELEKTROEĞİRME YÖNTEMİ İLE ÜRETİMİ

EMULSION ELECTROSPINNING OF PVA NANOFIBERS CONTAINING HYPERICUM PERFORATUM OIL

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ABSTRACT: The aim of this study is to produce essential oil loaded nanofiber wound dressings from PVA, which is a biodegradable and biocompatible polymer, by emulsion electrospinning method. Hypericum Perforatum oil, known for its antibacterial, antioxidant and anti-inflammatory properties on skin disorders, was used and Kolliphor RH40 surfactant was added to emulsify oil in polymer solution. In order to examine the effect of surfactant amount on nanofiber morphology, surfactant was used at 2%, 4% and 6% (w/w) ratios. The effect of surfactant concentration on solution viscosity and fibre morphology were investigated by viscosity measurements and SEM analysis. To confirm the presence of Hypericum Perforatum oil in nanofibers structure FT-IR spectroscopy analysis were carried out. According to the results, the solution viscosity increased as the amount of surfactant increased. Additionally, it was observed that the average diameters of nanofibers, containing 2%, 4%, and 6% surfactant (w/w), increased compared to the pristine PVA nanofiber. In conclusion, the fiber morphology of Hypericum Perforatum oil-containing PVA nanofibers has been changed with the effect of surfactant.

Keywords: emulsion electrospinning, essential oil, wound dressing, nanofiber

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ÖZ: Bu çalışmanın amacı, biyobozunur ve biyouyumlu bir polimer olan PVA'dan emülsiyon elektro-eğirme yöntemi ile uçucu yağ yüklü nanolifli yara örtüleri üretmektir. Cilt rahatsızlıklarında antibakteriyel, antioksidan ve antienflamatuar özellikleri ile bilinen sarı kantaron (hypericum perforatum) yağı katkı maddesi olarak kullanılmış ve yağın polimer çözeltisi içinde emülsiyon haline getirilmesi için Kolliphor RH40 yüzey aktif maddesi eklenmiştir. Miktarının nanolif morfolojisi üzerindeki etkisini incelemek amacıyla yüzey aktif madde %2, %4 ve %6 (w/w) oranlarında kullanılmıştır. Yüzey aktif madde konsantrasyonunun çözelti viskozitesi ve lif morfolojisi üzerindeki etkisi viskozite ölçümleri ve SEM analizi ile araştırılmıştır. Nanolif yapısında sarı kantaron yağının varlığını doğrulamak için FT-IR spektroskopi analizi yapılmıştır. Sonuçlara göre, yüzey aktif madde miktarı arttıkça çözelti viskozitesinin arttığı gözlenmiştir. Buna ek olarak, çözelti viskozitesinin artması nanoliflerin ortalama çaplarının da artmasına neden olmuştur. Sonuç olarak, sarı kantaron yağı katkılı PVA nanoliflerinin lif morfolojisinin yüzey aktif maddenin etkisiyle değiştiği gözlenmiştir.

***Anahtar Kelimeler*:** emülsiyon elektro eğirme, uçucu yağ, yara örtüsü, nanolif

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1. INTRODUCTION

Essential oils and natural plant extracts can be alternatives to synthetic drugs due to their antibacterial, antioxidant, and anti-inflammatory properties [1]. Hypericum Perforatum (HP) oil, which has anti-inflammatory, antimicrobial, and antioxidant features, is widely used in the treatment of skin and infectious diseases, [2]. Nanofibers with drugs, essential oil or extracts can provide controlled release due to their large surface areas and maintain the moisture balance on the skin surface. Therefore, nanofibers which contain drugs and essential oils are widely used in wound dressing applications. In the emulsion electrospinning method, which is frequently used in the production of nanofibers containing essential oils, the stable emulsion is formed with the help of suitable surfactants.

Recently, HP oil (St. John's Wort), which has anti-inflammatory, antimicrobial, antioxidant and pain reliever properties, has started to attract attention as one of the most used medicinal plants in the world. It is studied in detail in the pharmaceutical industry due to its components such as hypericins, hyperforins and flavonoids. Hyperforin is the main ingredient that provides antibacterial properties and helps skin epithelialization [3]. HP oil, which is also used in alternative treatment methods in Turkey, has a wide range of applications in the treatment of different wounds and burns [4]. Suntar et al. reported that HP oil increases infection resistance, has an anti-inflammatory effect that will contribute to rapid healing of the wound by providing fibroblast migration and collagen deposition [5]. There are several studies about electrospun wound dressings with HP oil content. In one of these studies, PCL/Gelatin electrospun wound dressings with HP oil were formed and in vivo tests were done. Consistent with the literature, it has been stated that HP oil has significant effects on wound healing [2]. In another one, HP oil encapsulated in PEG fibers showed a controlled release for 14 days upon contact with the wound and it was stated that electrospun structures containing HP showed activity against gram positive and gram negative bacteria [6]. In the study published by Schempp et al on the use of HP in the treatment of AD, it was shown that the phloroglucin derivative hyperforin component of HP has a visible antibacterial property against Staphylococcus Aureus bacteria, which is one of the causes of AD [7]. In addition to electrospun surfaces, different dressing studies where HP oil was loaded were also carried out. Bolgen et al. fabricated HP oil-loaded chitosan cryogel surfaces and tested the antibacterial and antioxidant properties of these surfaces. As a result of this study, it was concluded that HP oil-loaded chitosan cryogel scaffolds with antimicrobial and antioxidant properties can be used as wound dressings for exudative and long-healing wounds in tissue engineering applications [8].

In electrospun wound dressing studies synthetic polymers such as polylactic acid (PLA), polyglycolic acid (PGA), polyvinyl alcohol (PVA), polycaprolactone (PCL), polyvinylpyrrolidone (PVP), poly lactic-co-glycolic acid (PLGA) is used due to their biocompatibility [9]. In the literature, there are various studies in which electrospun nanofibers were obtained by doping PVA nanofibers with essential oils such as cinnamon oil, thyme oil and Zataria Multiflora oil [10-12]. In this study, with a different approach, emulsion electrospinning was chosen as the nanofiber production method by adding Hypericum Perforatum oil to the PVA polymer with the aim of developing wound dressings that could provide controlled release by forming encapsulated oil in the nanofiber structure. Since there is no other study conducted with PVA and HP oil in the literature to the best of our knowledge, we focused on investigation of the fiber formation by using the surfactant at different ratios. It is aimed to observe the nanofiber diameter and uniformity depending on different surfactant ratios, and thus to determine suitable formulations for electrospun wound dressing.

2. MATERIALS AND METHODS

In this study, PVA was obtained from Sigma-Aldrich and distilled water was used as the solvent. Hypericum Perforatum oil was obtained from Zade Vital and Kolliphor® RH 40 (BASF) was used as the non-ionic surfactant. Before the electrospinning process, emulsions containing PVA, Hypericum Perforatum oil and Kolliphor® RH 40 were prepared. PVA was dissolved in distilled water at a rate of 12% (w/w) for 2 hours at 70 °C and solution containing 12% (w/w) PVA was used in all samples. Amount of HP oil added was 2% (v/v). Kolliphor® RH 40 (BASF) was used as the surfactant and in order to observe the effect of surfactant ratio on nanofiber structure, Kolliphor® RH 40 was added at 2%, 4%, and 6% (w/w) ratios to PVA- Hypericum Perforatum oil emulsion. Viscosity measurements of the emulsions were carried out prior to spinning and Brookfield DV-E Viscometer was used for the measurements. NanoSpinner NE300 model electrospinning device was used and the feed rate was set as 0.3 mL/hour. The system voltage was fixed at 25 kV, the distance was 15 cm and the roller speed was determined as 200 rpm. TESCAN - VEGA4 - W source, EDX Scanning Electron Microscope (SEM) was used for image analysis of nanofiber surfaces. Fibre diameters were measured with the ImageJ software by selecting 100 different fibres from SEM images. Fourier Transform Infrared Spectroscopy (FT-IR) measurements of the samples were made using PerkinElmer Spectrum100.

3. RESULTS AND DISCUSSION

The measured viscosities of each polymer solution and the content of the pristine PVA nanofiber and oil loaded PVA electrospun fibers (HP-C2, HP-C4, and HP-C6) are presented in Table 1.

Since the aim of the study has been to examine the effect of the amount of surfactant on the nanofiber morphology, the amount of surfactant was increased by keeping the amount of oil constant at 2% (v/v). The addition of surfactant to a polymer solution can affect the viscosity of solutions by modulating the polymer/ solvent and polymer/surfactant interactions via electrostatic, hydrophobic, and hydrogen bonding interactions [13]. In accordance with this, the results (Table 1) showed that the viscosity of the solution increased as the amount of surfactant in the polymer solution increased.

Table 1. The content and the viscosity values of polymer solutions for each sample group.

	PVA/solvent (w/w)	The amount of surfactant (w/w)	The amount of oil (v/v)	Viscosity (cP)
Pristine PVA	12%	-	-	91.5
HP-C2	12%	2%	2%	184.5
HP-C4	12%	4%	2%	270.8
HP-C6	12%	6%	2%	328.0

The SEM images at 20,000 magnification and fibre diameter distributions of electrospun fibres are shown in Figure 1. The viscosity of the solution is a parameter that has an effect on the morphological structure and average diameter of the nanofibers [14]. As shown in the SEM images and fibre diameter distribution graphs (Fig. 1), the diameters of the fibers have thickened due to the increased viscosity caused by the addition of surfactant at 2%, 4%, and 6% (w/w) ratios. According to the results, the average diameter of electrospun fibres were measured as 82 ± 17 nm, 131 ± 47 nm, 176 ± 59 nm, and 208 ± 105 nm for the pristine, HP-C2, HP-C4, and HP-C6 samples, respectively.

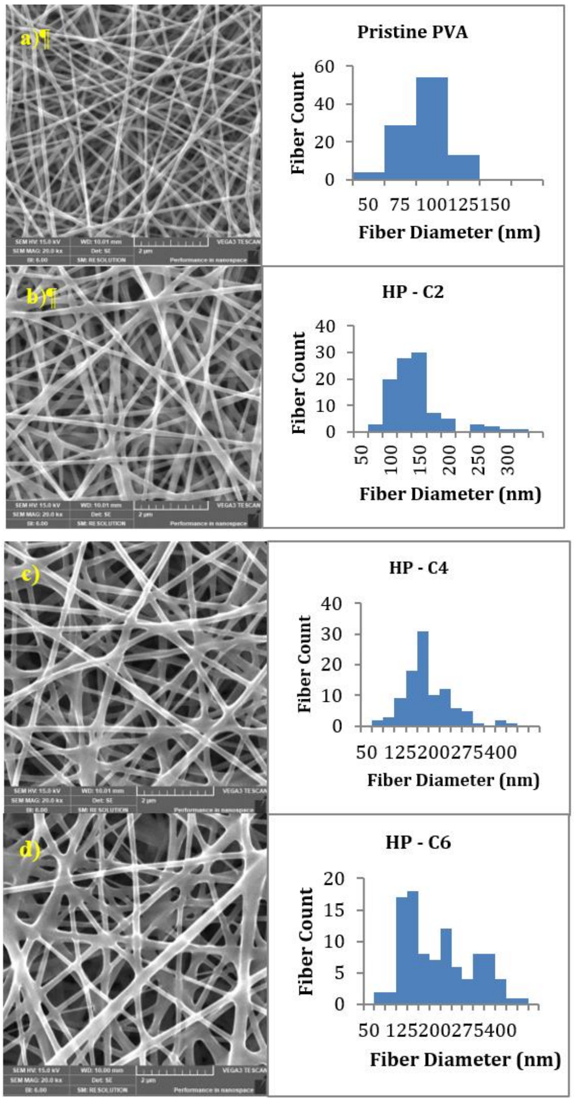

Figure 1. SEM images (at x20.000 magnifications) and fibre diameter distribution graphs of pristine PVA nanofiber (a), HP-C2 (b), HP-C4 (c), HP-C6 (d) samples, respectively.

In the literature, the increase in the diameter of the fibers has been accepted as an indicator of the encapsulation of more oil in the fiber [15]. In addition to the increase in fiber diameters, the fibers tended to coalesce and lose their fiber morphology in some parts (Figure 2). The reason for these morphological changes can be attributed to the increased amount of surfactant and viscosity.

As may be seen in Figure 3, in the FT-IR analysis, peaks similar to those of Hypericum Perforatum oil were observed for the PVA nanofibers with oil content. Peaks due to C-H stretching were observed in the region of 2850-3000 cm^{-1} in both HP oil and PVA nanofiber doped with oil. This is promising since it indicates that PVA nanofibers contain Hypericum Perforatum oil in their structure.

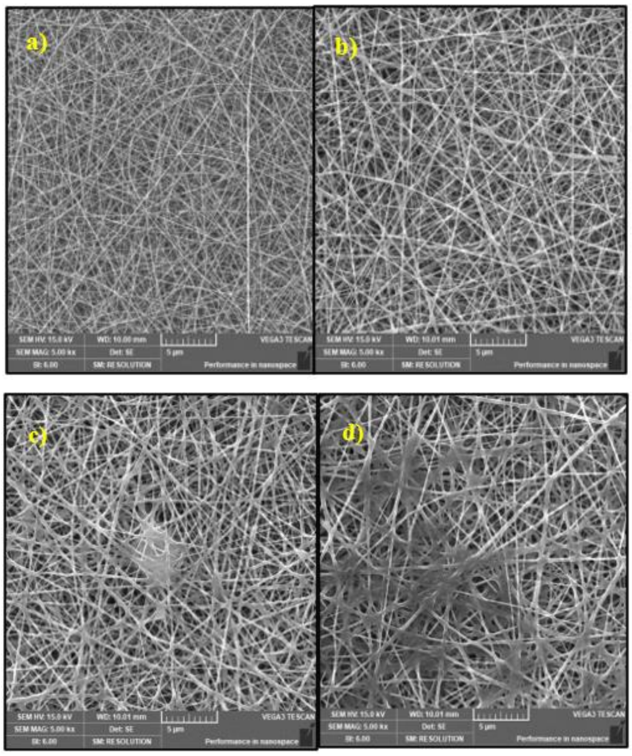

Figure 2. SEM images of pristine PVA nanofibers (a), HP-C2 (b), HP-C4 (c), HP-C6 (d) samples at x5.000 magnifications.

4. CONCLUSION

Nanofibers obtained by doping with HP oil, which is known to be effective in wound healing, have the potential to release oil in a controlled manner for skin treatments. In this study, nanofibers containing HP oil, which has antibacterial, antioxidant and anti-inflammatory properties, were produced from PVA polymer by emulsion electrospinning method. The aim of the study is to

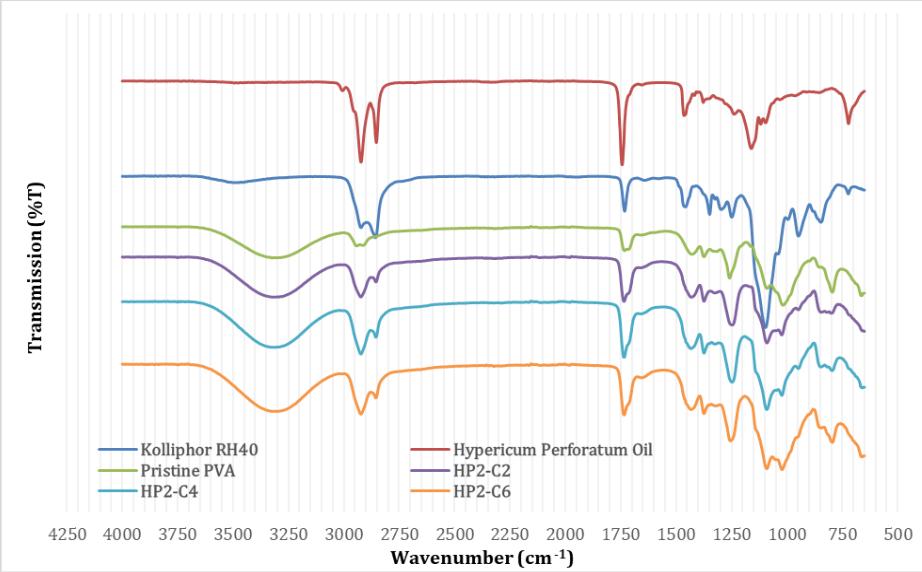

Figure 3. FT-IR Spectrum of nanofibers, Kolliphor RH40, and Hypericum Perforatum Oil

evaluate the effect of the amount of the surfactant added on the nanofiber structure. In order to create an emulsion between the oil phase and the water phase in the PVA solution, Kolliphor RH40 was used as the surfactant. Viscosity measurements done and SEM images were taken to examine the effects of surfactant concentration on both solution viscosity and fiber morphology. The results showed that as the amount of surfactant increased, fiber diameters and diameter distribution increased. On the other hand, in the FTIR measurements C-H stretching peaks in the 2850 - 3000 region of the HP oil were also observed in the PVA – HP Oil fibers. FTIR results indicate that HP oil was successfully incorporated into the fibre structure.

The effect of the surfactant on fiber morphology was investigated for further studies that aim to provide controlled release of essential oils trapped in the fiber by emulsion electrospinning method, especially during the treatment of skin disorders. In the future, the amount of oil in the solution will be increased and studies will be carried out on the maximum amount of oil that the fibers can carry without deteriorating their structure. By this means, depending on the amount of oil, the antimicrobial activities of nanofibers and their contribution to wound healing can be evaluated. In addition to that, by the analysis of the hypericin and other components contained in the HP oil, the release of the active substance and antimicrobial effect of oil-loaded nanofibers can be evaluated in future studies.

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REFERENCES

1. Mele E., (2020), *Electrospinning of essential oils,* Polymers, 12(4): 908.
2. Güleken Z, Depciuch J, Ege H, İlbay G, Kalkandelen C, Ozbeyli D, ... & Kuruca S.E., (2021), *Spectrochemical and biochemical assay comparison study of the healing effect of the Aloe vera and Hypericum perforatum loaded nanofiber dressings on diabetic wound,* Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 254: 119639.
3. Saddiqe, Z., Naeem, I. and Maimoona, A., (2010), *A review of the antibacterial activity of Hypericum perforatum L,* Journal of ethnopharmacology, *131*(3), pp.511-521.
4. Güneş, S., & Tıhmınlıoğlu, F. (2017), *Hypericum perforatum incorporated chitosan films as potential bioactive wound dressing material,* International journal of biological macromolecules, *102*, 933-943.
5. Süntar, I.P., Akkol, E.K., Yılmazer, D., Baykal, T., Kırmızıbekmez, H., Alper, M. and Yeşilada, E., (2010), *Investigations on the in vivo wound healing potential of Hypericum perforatum L,* Journal of ethnopharmacology, 127(2), pp.468-477.
6. Eğri, Ö. and Erdemir N., (2019), *Production of Hypericum perforatum oil-loaded membranes for wound dressing material and in vitro tests*, Artificial cells, nanomedicine, and biotechnology, 47(1): 1404-1415.
7. Schempp, C.M., Windeck, T., Hezel, S. and Simon, J.C., (2003), *Topical treatment of atopic dermatitis with St. John's wort cream–a randomized, placebo controlled, double blind half-side comparison.* Phytomedicine, 10, pp.31-37
8. Bölgen, N., Demir, D., Yalçın, M.S. and Özdemir, S., (2020), *Development of Hypericum perforatum oil incorporated antimicrobial and antioxidant chitosan cryogel as a wound dressing material.* International Journal of Biological Macromolecules, 161, pp.1581-1590
9. Jain R., Shetty S, & Yadav KS., (2020), *Unfolding the electrospinning potential of biopolymers for preparation of nanofibers,* Journal of Drug Delivery Science and Technology, *57*, 101604.
10. Wen P, Zhu DH, Wu H, Zong MH, Jing YR., Han SY., (2016), *Encapsulation of cinnamon essential oil in electrospun nanofibrous film for active food packaging,* Food Control, 59: 366-376.

11. Ardekani NT, Khorram M, Zomorodian K, Yazdanpanah S, Veisi H, & Veisi H., (2019), *Evaluation of electrospun poly (vinyl alcohol)-based nanofiber mats incorporated with Zataria multiflora essential oil as potential wound dressing,* International journal of biological macromolecules, 125: 743-750.

12. Koushki P, Bahrami SH, Ranjbar-Mohammadi M., (2018), *Coaxial nanofibers from poly (caprolactone)/poly (vinyl alcohol)/Thyme and their antibacterial properties,* Journal of industrial textiles, 47(5): 834-852.

13. Kriegel C, Kit KM, McClements DJ, Weiss J., (2009), *Influence of surfactant type and concentration on electrospinning of chitosan–poly (ethylene oxide) blend nanofibers*, Food Biophysics, 4(3):213-28.

14. Liao N, Unnithan AR, Joshi MK, Tiwari AP, Hong ST, Park CH, Kim CS., (2015), *Electrospun bioactive poly (ε-caprolactone)–cellulose acetate–dextran antibacterial composite mats for wound dressing applications,* Colloids and Surfaces A: Physicochemical and Engineering Aspects, 469:194-201.

15. Zhang C, Zhang H., (2018), *Formation and stability of core–shell nanofibers by electrospinning of gel-like corn oil-in-water emulsions stabilized by gelatin*, Journal of agricultural and food chemistry, 66(44):11681-90.