Electrospun Cactus Mucilage/Poly(vinyl alcohol) Nanofibers as a Novel Wall Material for Dill Seed Essential Oil (Anethum graveolens L.) Encapsulation: Release and Antibacterial Activities

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Journal:	*ACS Applied Materials & Interfaces*
Manuscript ID	Draft
Manuscript Type:	Article
Date Submitted by the Author:	n/a
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KEYWORDS: Polyvinyl alcohol, cactus mucilage, dill seed essential oil, emulsion electrospinning, nanofibers, antibacterial.

ABSTRACT: The goal of this work was to make a long-lasting carrier by making dill seed (*Anethum graveolens* L.) essential oil-loaded electrospun nanofibers from cactus mucilage (CM) and polyvinyl alcohol (PVA). Continuous and homogeneous nanofibers with an average

diameter ranging between 158 ± 18 and 230 ± 26 nm were successfully electrospun from the CM/PVA blend solution and the CM/PVA/DSEO emulsion. Atomic force microscopy topographic images revealed that the electrospun nanofibers had a tubular morphology. The TGA curves of DSEO, CM, pure PVA, and electrospun nanofibers show successful chemical interactions between the polymers used and the essential oil. The water contact angle results suggest that the manufactured nanofibers are hydrophilic. CM/PVA consistently achieves a remarkable encapsulation efficiency of 100% DSEO. The electrospun nanofibers allowed for controlled release of free and encapsulated DSEO, with sustained long-term oil release. The agar disk diffusion technique was used to study the antimicrobial activity of electrospun nanofibers and nanofibers containing DSEO against Gram-positive and Gram-negative bacteria. Electrospun nanofibers containing DSEO exhibited bacteriostatic and bactericidal activities against foodborne pathogenic bacteria (*S. aureus* and *P. aeruginosa*) with MICs of 2.5 mg/mL and MBCs of 5 mg/mL. The electrospun nanofibers based on carbohydrates loaded with DSEO demonstrate potential as an active internal coating for food packaging and biomedical applications.

1. INTRODUCTION

Electrospinning is one of the progressing technologies in nanofiber production. It is a versatile and potentially useful technique for creating micro- and nanoscale fiber mats with effective morphological and mechanical properties.[1] Electrospinning is a current and simple technique that uses high electric fields to stretch polymer solutions or melts to produce long-lasting and continuous fibers with 10–1000 nm diameters.[2,3] Solution electrospinning is more commonly employed for the production of nanofibers than melt electrospinning. It is mainly used because of its low cost and potential for use in laboratory research and nanofibers' industrial production.[4] For solution electrospinning, various kinds of organic polymers dissolved in solution form are adopted to produce nanofibers and nonwovens. Electrospinning has different factors that affect polymer solutions, such as polymer molecular weight, viscosity, concentration, solvent volatility, conductivity, and surface tension, and the electrospinning process, such as spinning voltage, spinning flow rate, needle tip to collector distance, temperature, and humidity.[5] Electrospun nanofibers can be produced based on synthetic organic polymers and blends of solutions with natural polymers, including proteins, nucleic acids, and polysaccharides.[6,7] The use of biopolymers in electrospinning has attracted great attention due to their large specific surface

area, good biocompatibility, biological activity, biodegradability, and non-toxicity, as well as their diversified potential in technical and biomedical applications such as functional textiles, filtration membranes, tissue engineering scaffolds, wound healing, drug delivery, cosmetics, essential oil encapsulation, and active packaging.[6,8−11] Despite recent progress in the conventional electrospinning process, technological challenges exist to produce core–shell nanofibers using emulsion electrospinning. It is a novel and simple method that has rapidly developed and has received increasing interest as the process is considered more stable for encapsulating therapeutic and bioactive compounds and essential oils within a single carrier.[12] In the electrospinning process, selecting a suitable encapsulation material is important and is widely studied depending on the nature of the uses for food, biomedicine, and other applications. In this way, materials from renewable and alternative resources are used as appropriate wall materials for emulsion electrospinning, such as biopolymers like proteins and polysaccharides, including mucilage, cellulose and derives, starch, chitosan, etc., and biocompatible polymers essentially the polyvinyl alcohol, polyethylene oxide, and polycaprolactone.[12−17] Polyvinyl alcohol is a non-ionic hydrolysis derivative of polyvinyl acetate. It is a highly hydrophilic semi-crystalline polymer with excellent mechanical properties.[18] PVA is commonly used alone or blended with

natural polymers to produce electrospun nanofibers. The emulsion-based electrospun nanofibers are made from blended polymers according to their type and specific uses.[19] Moreover, the emulsion electrospinning process is proposed as an effective way to encapsulate the essential oil in fiber-forming materials. The encapsulation efficiency tends to be 100%. Electrospun nanofibers have proven their suitability as materials for the controlled release of essential oils.[20] This method is based on the electrospinning of stable biopolymeric emulsions, which can be made by stabilizing water-in-oil or oil-in-water emulsions with an emulsifier or surfactant. These emulsions can then be electrospun as spinning solutions. Meanwhile, other essential oils have been loaded into electrospun fibers, like thyme essential oil,[21] costmary essential oil,[22] cinnamon essential oil,[23] orange essential oil,[20] cabreuva essential oil[15] and cumin essential oil.[24] In this case, DSEO was microencapsulated through emulsion electrospinning, and, for this reason, CM is a fascinating and interesting core-shell material for emulsion electrospinning. Additionally, PVA can be blended with mucilage and used as a fibrous carrier for essential oils. The oil-in-water emulsion was prepared using a high-speed homogenization process.

Essential oils have been widely exploited for their biological properties, mainly as antimicrobials in the food, pharmaceutical, and biomedical fields. Encapsulation of essential oils

is necessary to protect their functional properties because they have the characteristics of fast release and a short duration of action. Emulsion electrospinning is a promising technique for essential oil encapsulation to improve its sustained-release performance.[25] Essential oils from dill seeds are the most valuable medicinal seed spice crops of the Apiaceae.[26] It is one such annual aromatic herb, belonging to the family Umbelliferae.[27] DSEO is a potential inhibitor of bacterial and fungal.[27−29] Further, it was found that the essential oils of *Anethum graveolens* L. seeds have been reported as a potential natural preservative in fish food due to their antioxidant and antimicrobial activities.[28]

The cactus, *Opuntia ficus-indica*, is a tree-like shrub that belongs to the *Cactaceae* family. It is a suitable carbohydrate source and can adapt to various environmental conditions.[30] The cactus biomass has been mainly used as a raw material for added-value materials with different applications, such as reinforcing agents for the production of composite materials,[31,32] papermaking[33,34] and wastewater treatment.[35] The cactus mucilage extracted from the cladode is a heteropolysaccharide of an anionic polyelectrolyte nature with a high molecular mass, mainly composed of L-arabinose, D-galactose, D-xylose, L-rhamnose, and α-D-(1 → 4) galacturonic acid.[36] CM exhibits an effective functional property due to the hydrogen bonding between

different functional and other polar groups. Cactus mucilage has been utilized in food as a thickener, in cosmetics, and in pharmaceuticals as a wall material for encapsulation processes[37–39] as well as in film-forming.[40]

To the best of our knowledge, there have been no previous reports of DESO loading into blended CM and PVA electrospun nanofibers. The DSEO was extracted from ground dill seeds using hydrodistillation, and the oil's chemical composition was identified using gas chromatography coupled to mass spectrometry. The CM was extracted with hot water and a sonication process from fresh cladodes and then precipitated with ethanol. The CM extracted using ethanol as the extraction solvent is considered non-toxic and safe for human and animal consumption. The spinning solution and emulsion were created by combining CM and PVA. The oil-in-water emulsion was carried out using DSEO as the oily phase and a CM/PVA blend solution as the aqueous phase. The pH, viscosity, conductivity, and surface tension of the obtained solutions were controlled. The polymer solutions were used to formulate neat PVA/CM, and loaded CM/PVA/DSEO electrospun nanofibers. They were considered an alternative to producing nanostructures and encapsulating the essential oil during fiber formation by the

electrospinning method. The electrospun fiber morphology, structural and thermal properties, and water contact angles were determined. The DSEO release was controlled.

2. EXPERIMENTAL SECTION

2.1. Extraction and characterization of essential oil from dill seeds.

Dill seed was collected from the El-Guettar oasis, near Gafsa, in the south-west of Tunisia. The extraction of the dill seed essential oil was carried out by the hydrodistillation method. Approximately 100 g of the powdered dill seeds (≥45 μm) were immersed in 2 L of distilled water, and the resulting mixture was hydrodistilled for 3 h (until there was no more water to evaporate). The DSEO were separated, dried with anhydrous sodium sulfate, and stored in a dark glass vial at 4 ˚C for further experiments. The chemical characterization of DSEO was carried out by gas chromatography/mass spectrometry (GC/MS, GC 7890A, MS 5975C, Agilent, Santa Clara, CA, USA). About 0.01 g of dill seed essential oil was dissolved in 1 ml of ethyl acetate, and then this solution was injected into a capillary column (Agilent, 0.25 mm × 30 m). The conditions used to perform the GC/MS analysis were as follows: the injector was at 280 ˚C with the split/splitless method in split mode (10:1); the carrier gas washes helium with a flow rate of 0.7 mL/min; the temperature ramp started at 50 ˚C and was increased by 10 ˚C/min to 120 ˚C

and maintained for 5 min; in the second stage of the ramp, the temperature was increased from 10 ˚C/min to 280 ˚C and maintained for 8 min; finally, in the third stage, the temperature was increased by 10 ˚C/min to 300 ˚C and kept constant for 2 min. The identification of all components was studied using a relative peak area percentage. An attempt to identify the compounds was made based on the comparison of their relative retention times and mass spectra. The confirmation of the identified components of the dill seed essential oil was carried out with the National Institute of Standards and Technology (NIST). Essential oil components were determined using the NIST library.

2.2. Extraction of cactus mucilage

CM was extracted from fresh cladodes harvested from the El-Guettar oasis, near Gafsa in the south-west of Tunisia, and cleaned and washed with distilled water. Fresh cladode pieces (1×1×1 cm^3) were homogenized with distilled water (1:1 w/v), soaked overnight, and then boiled at 80 ˚C for 40 minutes under mechanical stirring. After cooling, the mixture was put in an ultrasonic bath for 40 minutes at 40 ˚C. Thus, the mixture was filtered three times through a nylon filter to separate the liquid from the solids and centrifuged for 20 minutes at 4000 rpm at 25 ˚C. The liquid was recovered and precipitated with ethanol (96% purchased from Sigma-Aldrich) at a

ratio of 1:3 (v:v). Precipitation was performed at 4 ˚C overnight. The CM was washed with pure ethanol (99% purchased from Sigma-Aldrich) and acetone (99% purchased from Sigma-Aldrich), dried at 45 ˚C for 24 hours, and then stored in bottles and dark glass. The extraction yield of CM was 22.8 ± 2.4%.

2.3. Preparation of spinning solution

The CM solution was prepared by introducing 2 g of dried CM into 100 mL of distilled water. The mixture was homogenized using magnetic stirring overnight at room temperature and humidity. The concentration of PVA (Mw: 145000 g/mol, bought from Sigma-Aldrich) was set at 15 wt%, and was dissolved at 70 ˚C for 3 hours with magnetic stirring to make a thick solution that could be electrospun. After cooling, the CM/PVA mixture was prepared in a ratio of 80:20 (w/w) under magnetic stirring for 30 min. The oil-in-water emulsion was made with a dispersed oil phase of 4% DSEO (w/w, oil/polymeric aqueous solution) and a continuous phase of CM/PVA solution. The Tween 80 was used as an emulsifier (1.2 wt% of the oil mass). The mixture was then vigorously dispersed with an ultraturrax high-speed homogenizer (Ika T 25 basic, France) at room temperature (≈20 ˚C) and 11000 rpm for 15 min to better stabilize the oil droplets in the CM/PVA solution.

2.4. Electrospinning

The electrospinning of CM/PVA and CM/PVA/DSEO mixtures was performed using Esprayer ES-2000S2A equipment (Fuence Co., Ltd., Tokyo, Japan) at 25˚C and a relative humidity of 34%. A metallic electrode plate wrapped with aluminum foil was placed 15 cm (depending on the sample) away from the needle (Nordson Stainless Steel Tips, 18 Gauge; inner diameter = 0.84 mm; outer diameter = 1.27 mm). High voltage (30 kV) was applied between the needle and the electrode plate while the mucilage-blend solution flowed with a latency of 20 μL/min from the needle to collect the electrospun product on the aluminum foil. The electrospun products on the aluminum foils were dried at room temperature in desiccators. Figure 1 shows the scheme for the preparation of electrospun nanofibers.

2.5. Characterization of solutions

The conductivity and pH of the different samples were measured using a Seven Easy™ S30 conductivity meter, Belgium, and a Proviteq pH meter, France, respectively. The viscosity of the different solutions and emulsions was measured at 20 ˚C using a rotational rheometer with a 50-mm cone-plane measurement system (Anton Paar, PHYSICA MCR302 viscometers). This

makes it possible to measure the variation of viscosity (η) as a function of time and the speed of shear ($\dot{\gamma}$) from 10^{-3} to 10^{3} s^{-1}.

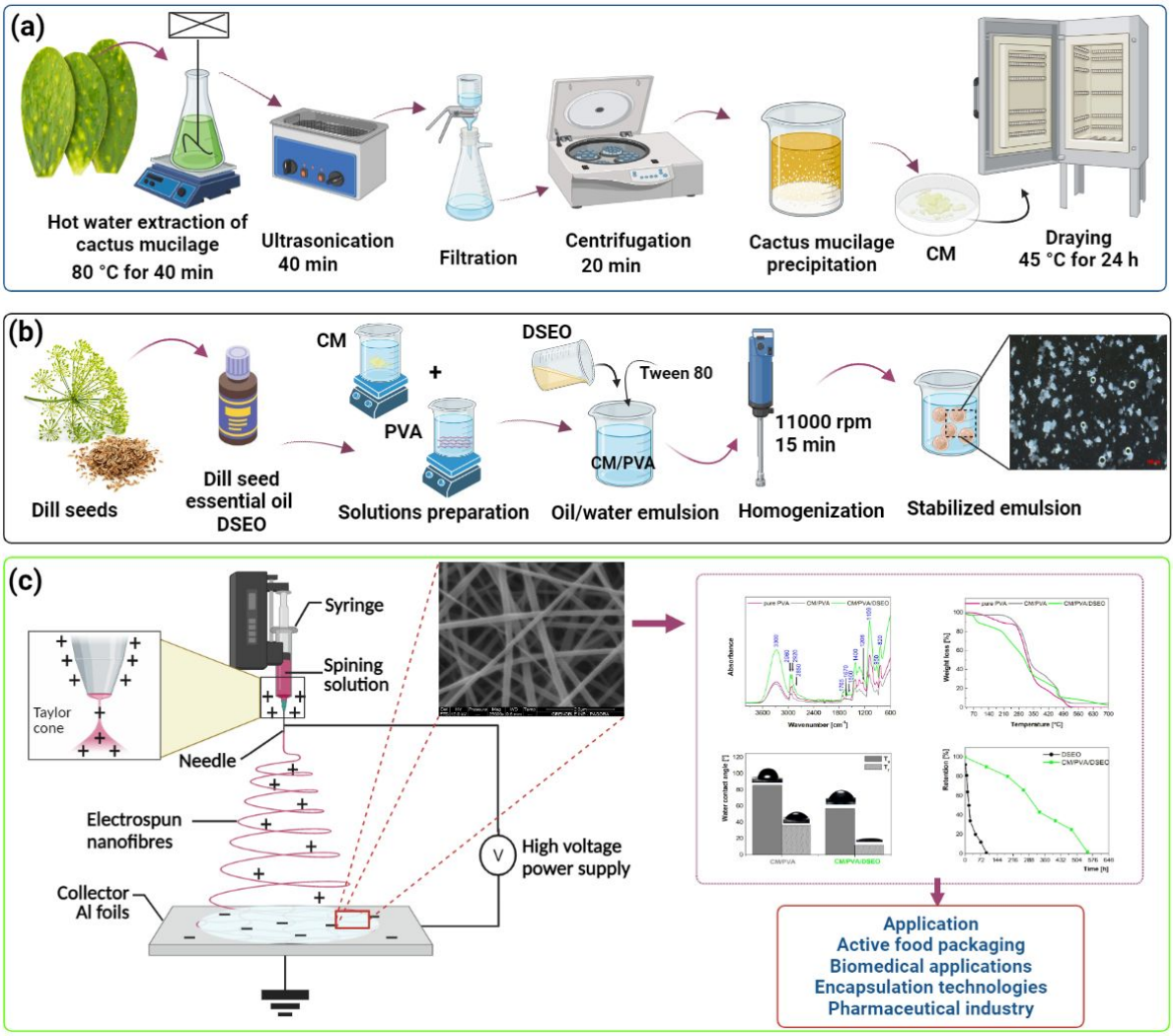


Figure 1. Schematic illustration of procedures used to produce electrospun nanofibers: (a) cactus mucilage extraction, (b) spinning solution and emulsion preparations and (c) electrospun nanofibres formulation.

2.6. Morphological analysis

The morphology of the prepared emulsion was observed by optical microscopy. The morphology of electrospun nanofibers was studied by scanning electron microscopes (SEM) and atomic force microscopy (AFM). An optical microscope, the Axio Imager M1 (Carl Zeiss, Munich, Germany), was operated in transmission mode, and the emulsion sample was observed at a diluted concentration. At least 10 images were taken. The electrospun nanofibers on the aluminum foil were coated with a ca. 4-nm-thick layer of gold/palladium (Au/Pd) before scanning electron microscopy observation. The scanning electron microscopy was performed using a Quanta 250 FEG (Thermo Fischer) device equipped with a CCD camera with a resolution of 16 bits, and the accelerator voltage was fixed at 10 kV. Five micrographs for each sample were captured at different positions. The topography of electrospun nanofibers was characterized using multimodal atomic force microscopy (DI, Veeco, Instrumentation Group, Plainview, NY, USA). Samples (0.2×0.2 cm^2) were deposited on a mica plate disc and then analyzed, and representative micrographs have been recorded and selected for results and discussion.

2.7. Fourrier transmission infrared analysis

The infrared spectra of samples were carried out on an Elmer Paragon 100 spectrometer using an attenuated total reflectance (ATR) module composed of diamond crystals. The measurements were recorded between 600 and 4000 cm^{-1}, with a resolution of 4 cm^{-1} and 20 scans.

2.8. Thermal analysis

The thermogravimetric analysis (TGA) of all samples was performed using an STA-6000 thermogravimetric analyzer (Perkin Elmer Instruments, Villebon-sur-Yvette, Grenoble, France). Approximately 7 to 10 mg of sample were dynamically scanned between 30 and 700 ˚C at 10 ˚C/min under an oxygen stream with a flow rate of 20 mL min^{-1}.

2.9. Contact angle

Water contact angle measurement was performed with a Theta Flex optical tensiometer (Biolin Scientific) using 5 µL water drops. The contact angles were recorded at initial drop contact on the surface of the specimen (T_0) and after 20 s of drop contact on the surface (T_f). Three measurements were performed for each sample at 20 ˚C.

2.10. Encapsulation efficiency

According to Yang et al.,[41] the encapsulation efficiency (EE) of DSEO loaded into CM/PVA nanofibers was studied by measuring the amount of oil located on the surface of electrospun nanofibers (non-encapsulated oil). 60 mg of the electrospun fibers were put in hexane (7 mL) for 1 min with handshaking to remove the surface oil. The absorbance of the obtained liquid was evaluated at a wavelength of 278 nm using a UV/vis spectrophotometer. The concentration of dill seed essential oil in hexane was determined using a calibration curve (R^2=0.99) obtained by dissolving different amounts of essential oil in hexane. The encapsulation efficiency was calculated as follows:

$$EE = \frac{T - F}{T} \times 100$$

where, T was the total amount of dill seed essential oil, F was the free amount of dill seed essential oil in the collection solution.[42]

2.11. Essential Oil release

The release of free and encapsulated DSEO was controlled according to the method of Gómez-Mascaraque et al.,[43] with some modifications. 2 mg of electrospun nanofibers were immersed in 7 mL ethanol and slowly stirred at room temperature. An aliquot of 1.5 mL solution was

removed and measured at a wavelength of 220 nm using a UV/vis spectrophotometer during specific intervals of time. The same volume as that of the aliquot (1.5 mL) was replaced with fresh ethanol at each interval.

2.12. Antibacterial activity of electrospun nanofibers

2.12.1. Bacterial strains and inoculums preparation

To assess the potency of each sample's antibacterial properties, we tested them against five bacterial strains that cause food poisoning. These strains included two types of Gram-positive bacteria (*Staphylococcus aureus* and *Bacillus cereus*) and three types of Gram-negative bacteria (*Escherichia coli*, *Salmonella typhi*, and *Pseudomonas aeruginosa*). The bacterial strains were provided from the culture collection of the Department of Botany and Microbiology at the Faculty of Sciences of Gafsa. Each bacterial strain was subcultured overnight at 35 ˚C on Mueller-Hilton agar slants. Bacterial growth was harvested using 5 mL of sterile saline water; its absorbance was adjusted to 580 µm and diluted to a viable cell count of 10^7 CFU/mL using a UV/vis spectrophotometer.

2.12.2. Antibacterial activity

The disk diffusion method was used to evaluate the antimicrobial activity of the produced electrospun nanofibers. The samples were loaded over sterile filter paper discs (8 mm in diameter) to obtain a final concentration of 10 mg/disc. About 10 mL of Mueller-Hilton agar medium was poured into sterile Petri dishes (as a basal layer) followed by 15 mL of seeded medium previously inoculated with bacterial suspension (100 mL of medium/1 mL of 10^7 CFU) to attain 10^5 CFU/mL of medium. Sterile filter paper discs loaded with a concentration of 10 mg/mL of electrospun nanofibers were placed on top of Mueller-Hilton agar plates. Filter paper discs loaded with 10 μg gentamicin and 10 μL dimethyl sulfoxide (DMSO, 99.9% purchased from Sigma-Aldrich) were used as positive and negative controls, respectively. The plates were kept in the fridge at 5 ˚C for 2 h. The sample diffusions were incubated at 35 ˚C for 24 h. The presence of inhibition zones was measured by a Vernier caliper and recorded and considered an indication for antibacterial activity.

2.12.3. Determination of minimum inhibitory concentrations (MIC's) and minimum bactericidal concentrations (MBC's)

MIC is defined as the lowest concentration of the antimicrobial agent that inhibits microbial growth after 24 h of incubation. The electrospun nanofibers, which exhibit strong antibacterial

activity at 10 mg/mL were manipulated to determine their MIC using the disk diffusion method and evaluate their efficiency in controlling bacterial strains causing food poisoning diseases. Different concentrations of electrospun nanofiber extract (2.5, 5.0, 10.0, 12.5, and 15.0 mg/mL) were prepared separately and loaded in their requisite amounts over sterilized filter paper discs (8 mm in diameter). Mueller-Hilton agar was poured into sterile Petri dishes and seeded with bacterial suspensions of the pathogenic strains. The loaded filter paper discs with different concentrations of the electrospun nanofibers were placed on top of the Mueller-Hilton agar plates. The plates were kept in the fridge at 5 ˚C for 2 h. Then, the samples were incubated at 35 ˚C for 24 h. The inhibition zones were measured by a Vernier caliper and recorded against the concentrations of the electrospun nanofibers.

For the MBC's, streaks were taken from the two lowest concentrations of the electrospun nanofibers plates exhibiting invisible growth (from the inhibition zone of MIC plates) and subcultures onto sterile Tryptone soya agar (TSA) plates. The plates were incubated at 35 ˚C for 24 h. Then, they were examined for bacterial growth corresponding to the sample concentration. MBC was taken as the concentration of samples that did not exhibit any bacterial growth on the freshly inoculated agar plates.

3. RESULTS AND DISCUSSION

3.1. Essential oil and solution properties

The extracted oil from dill seeds was pale yellow with a strong characteristic aroma and pleasant odor, and the yield was 3.50 ± 0.8% (v/w). The yield of DSEO extraction is higher than that of essential oils extracted from dill seeds planted in India, varying between 1.71 and 2.06%.[26] The DSEO constituents were identified with GC/MS analysis.

Table 1. Chemical composition of essential oil from dill seeds obtained from Gas chromatography-mass spectrometry analysis.

Peak No.	Compound	Content (%)
1	3,4,4-Trimethyl-3-pentanol	tr
2	Thiocyanic acid, ethyl ester	0.33
3	Camphene	0.43
4	3–Carene	tr
5	β–Phellandrene	tr
6	β–pinene	tr
7	δ–Carene	tr
8	α–Phellandrene	0.42
9	β–Cymene	0.32

10	d–Limonene	36.3
11	γ-Terpinene	2.2
12	Terpinolene	0.46
13	Camphor	0.87
14	Dihydrocarvone	2.6
15	Carvone	13.1
16	Piperitone	16.4
17	Apiol	25.5

tr: trace, relative content < 0.10%.

According to the results in Table 1, 17 components represented 98.93% of the total oil. The major components detected in the oil were d-limonene (36.3%), apiol (25.5%), piperitone (16.4%), and carvone (23.49%). *d*-limonene is a constituent extensively present in essential oils, already identified in commercial DSEO studied by Noumi et al.[44] The obtained results are in accordance with the literature.[27,29,45,46]

The pH, conductivity, viscosity, and surface tension of prepared solutions and emulsions were determined, and the results are listed in Table 2. The pH value of ≈ 5 indicates that prepared solutions and emulsions are slightly acidic. The viscosity of pure PVA (15% w/v) and the CM/PVA/DSEO emulsion was higher than that of the CM/PVA solution (Table 2).

Table 2. pH, conductivity, viscosity and surface tension of polymeric solutions and emulsion.

Solution	pH	Conductivity (mS/cm)	Viscosity ($mPas^{-1}$)	Surface tension (mN/m)
CM (2% *w/v*)	5.2	5.8 ± 0.3	8.5± 0.8	24.2± 0.1
PVA (15% *w/v*)	5.3	42.5 ± 3.2	848.2± 1.2	50.8± 0.4
CM/PVA	5.3	113.2 ± 0.9	421.4± 0.6	46.6± 0.6
CM/PVA/DSEO	5.3	126.5 ± 1.1	627.4± 0.5	26.5± 0.2

The viscosity of the produced polymeric solutions is acceptable to form nanofibers, but a low viscosity of the polymeric solution leads to spindle-shaped beads instead of nanofibers.[16] Also, the high viscosity of a polymeric solution can influence the degree of chain entanglement between polymers and generate large nanofibers. So, the viscosity can reach a critical point, and the nanofiber is generated.[16] In this study, the viscosity of polymeric solutions and emulsions can be used for electrospun nanofibers. The viscosity results agree with those of Rafiq et al.,[47] who studied the viscosity of PVA/sodium alginate and PVA/sodium alginate loaded with cinnamon, clove, and lavender. The conductivity of the PVA solution increases with increasing carboxylic groups and anionic polyelectrolytes in mucilage polysaccharides and with additions of Tween 80 emulsifier and DSEO. The surface tension of the pure PVA aqueous solution was 50.8 mN/m,

which was higher than the surface tension of CM and oil-containing solutions, which were 46.6 and 26.5 mN/m, respectively. It has been observed that adding CM polysaccharides and DSEO decreases the surface tension. This reduction may be due to the polysaccharide chains' reactivity, particle size distribution, and strong inter-polymer interactions between emulsion, surfactant, and polysaccharide macromolecules.[48]

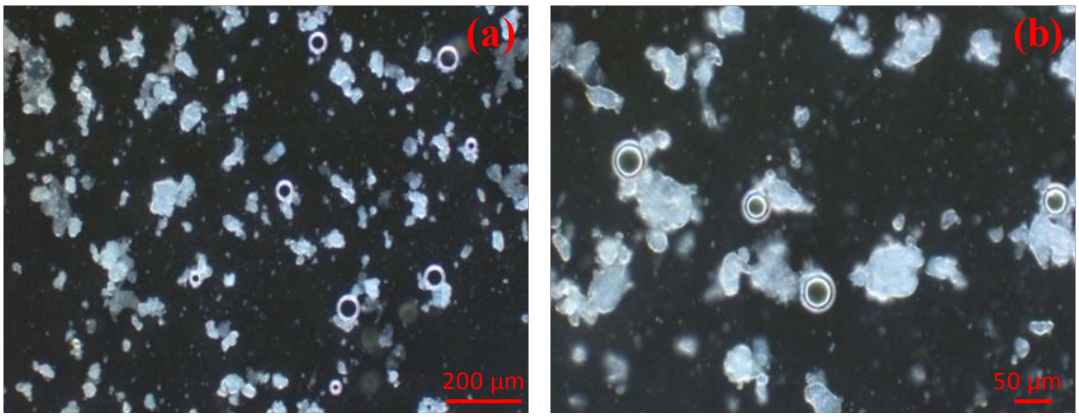


Figure 2. Optical microscopic images of an oil-in-water emulsion of DSEO/CM/PVA: (a) scale bars corresponds to 200 µm and (b) scale bars corresponds to 50 µm.

Figure 2 shows the optical microscopy images of the oil-in-water emulsion of DSEO in CM/PVA aqueous solution that were taken right after they were made. The O/W emulsion images show that the oil droplets have been encased as separate spherical globules with a thick shell (Figure 2b). These globules are polydispersed with different diameters, which shows that there are no agglomerates. The average diameter of microcapsules and droplets is between 1± 0.4

μm and 50 ± 1.2μm. The emulsification step involves a complex interaction between the oil and the CM/PVA aqueous phase and produces, with Tween 80, a stable emulsion. The CM/PVA/DSEO emulsion is more stable than the PVA/thyme essential oil emulsion prepared for emulsion electrospinning.[17]

3.2. Fiber morphology and topography

SEM micrographs of electrospun nanofibers are shown in Figure 3. Electrospinning CM/PVA and CM/PVA/DSEO emulsions made smooth, uniform nanofibers that were not woven and didn't have any microbeads.

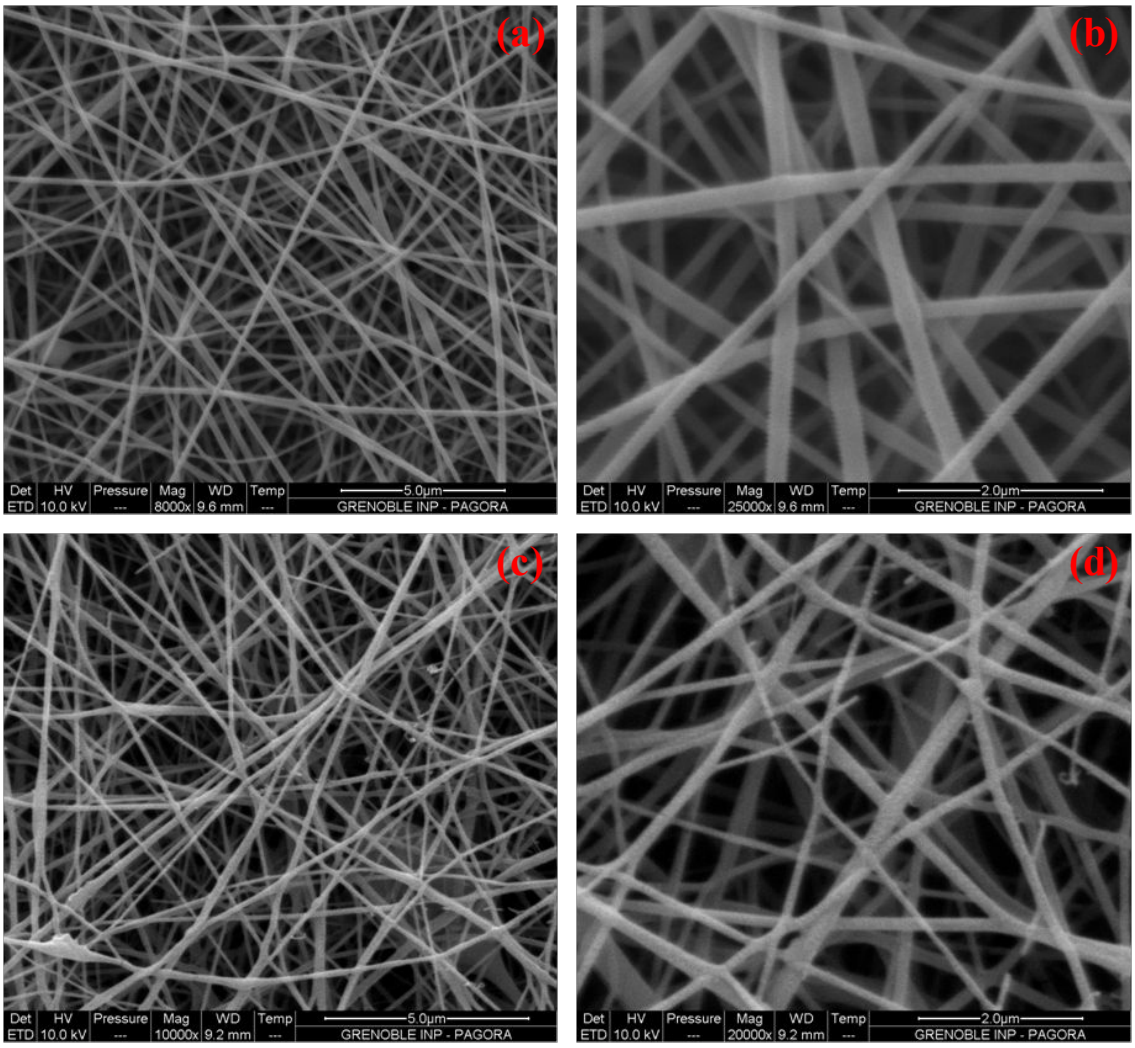


Figure 3. SEM micrographs of: (a-b) CM/PVA electrospun nanofibres [a: 8000× and b: 25000×] and (c-d) CM/PVA/DSEO electrospun nanofibres [c: 10000× and d: 20000×].

The CM/PVA electrospun nanofibers had an average diameter of 158±18 nm, whereas that of the loaded electrospun fibers with essential oil was slightly larger, *i.e.* 230 ±26 nm. The increase in diameter of CM/PVA/DSEO emulsion electrospun nanofibers is probably due to the essential oil encapsulation and incorporation into the nanospun nanofibers. Furthermore, emulsion

electrospinning produced bead-free fibers. In general, the morphology of electrospun fiber is strongly affected by the viscosity and conductivity of the spinning polymer emulsion, solution, or melt. The large values of viscosity and electrical conductivity in emulsion, compared to CM/PVA solution (Table 2), improve electrostatic interactions and other types of colloidal interactions like van der Waals, steric, depletion, and salt-induced attraction between macromolecules of polymeric solution. The effects of hydrogen bonding and interactions between polymers lead to broad and uniform nanospun without bead defects during electrospining.[49] Similar morphology has been observed in many other electrospun nanofibers loaded with different sources of essential oil, such as PVA/cinnamon essential oil/*β*–cyclodextrin electrospun nanofibers,[50] Polyvinylpyrrolidone/cinnamon essential oil,[23] PVA/gelatin nanospun loaded with thyme essential oil,[48] avocado oil in ultrafine zein nanofibers,[51] electrospun nanofibers of PVA/chitosan functionalized with cabreuva essential oil,[15] zein electrospun nanofibers loaded with cumin essential oil,[24] and costmary essential oil loaded salep-PVA electrospun.[22]

AFM is a useful tool for studying the topography of the electrospun nanofibers to further confirm that they are made from CM-blend PVA and emulsion. It can be seen that the AFM images

(Figure 4) are in good agreement with the obtained SEM micrographs in terms of fiber structures. The topographical surfaces show the tubular morphology of all electrospunnanofibers as reported elsewhere.[24]

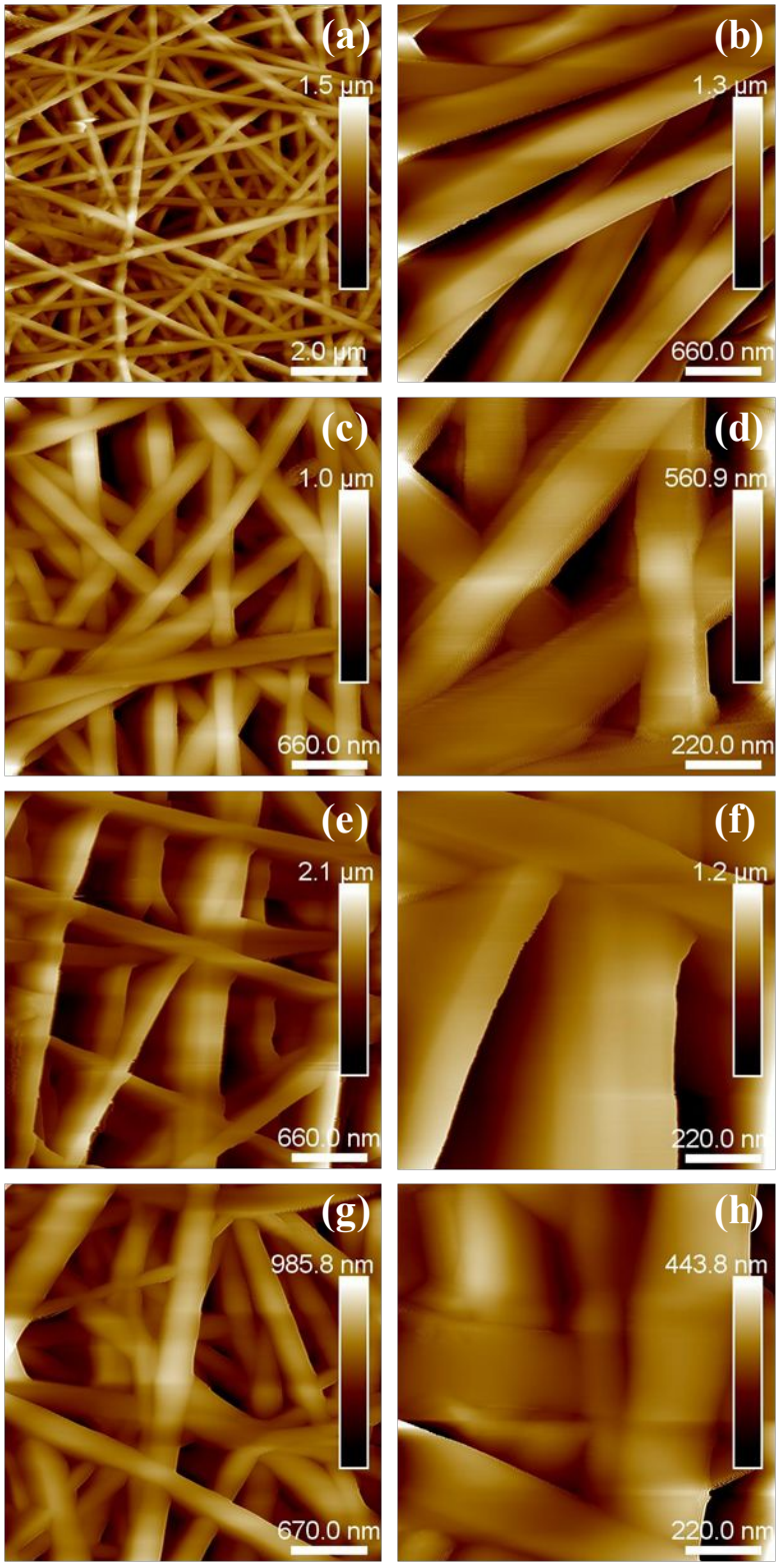

(a)
1.5 µm
2.0 µm
(b)
1.3 µm
660.0 nm
(c)
1.0 µm
660.0 nm
(d)
560.9 nm
220.0 nm
(e)
2.1 µm
660.0 nm
(f)
1.2 µm
220.0 nm
(g)
985.8 nm
670.0 nm
(h)
443.8 nm
220.0 nm

Figure 4. AFM images of the surface topography of: (a–d) CM/PVA electrospun nanofibres [a: scale bars 2 μm, b: scale bars 660 nm, c: scale bars 660 nm and d: scale bars 220 nm] and (e–h) CM/PVA/DSEO electrospun nanofibres [e: scale bars 660 nm, f: scale bars 220 nm, j: scale bars 670 nm and h: scale bars 220 nm].

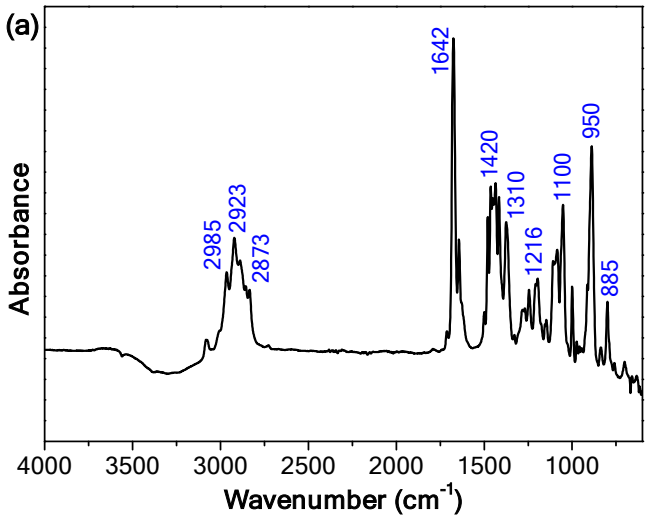
The topography image of CM/PVA electrospun nanofibers reveals a fibrillar structure with rough, laminated, and cross-linked nanospun fibers forming a nonwoven mat structure (Figure 4 a-d). The diameter of electrospun nanofibers increases when dill seed essential oil is added. It is confirmed that surface roughness increases with increasing electrospun fiber diameter.[52] The CM/PVA/DSEO regenerating fibers with encapsulated oil seem larger with entangled edges. Similar topography has been reported in other electrospun fabrics produced with polysaccharide-essential oil emulsions.[24,53]

3.3. ATR-FTIR

ATR-FTIR spectra of the extracted dill seed essential oil obtained by hydrodistillation (Figure 5a) show the presence of the characteristic vibration bands of the main components of DSEO. It can be observed that the peaks at 1310, 1216, 950, and 885 cm^{-1} are attributed to the double bonds in *d*-limonene as a major compound.[54,55] Also, a characteristic band at 1642 cm^{-1}

corresponds to the stretching band of C=C as a functional group in *d*-limonene, apiol, carvone, pinene, etc….[48] The bands are characteristic of the terpenes found in essential oils.[56] Other bands are observed between 3100 and 2850 cm^{-1}, related to $-CH$, $-CH_2$ and $-CH_3$ stretch vibrations.

The ATR-FTIR spectra of extracted CM are shown in Figure 5b, exhibiting a spectrum similar to that reported by other authors for cactus polysaccharide. The characteristic broadband at 3300 cm^{-1} corresponds to the stretching of $-OH$ groups of the polysaccharides in mucilage.[57] The small band situated at 2900 cm^{-1} is attributed to the $-CH_2$ group vibration of the pyranose backbone.[58] The band occurring at 1763 cm^{-1} is related to the C=O bonds.[59]



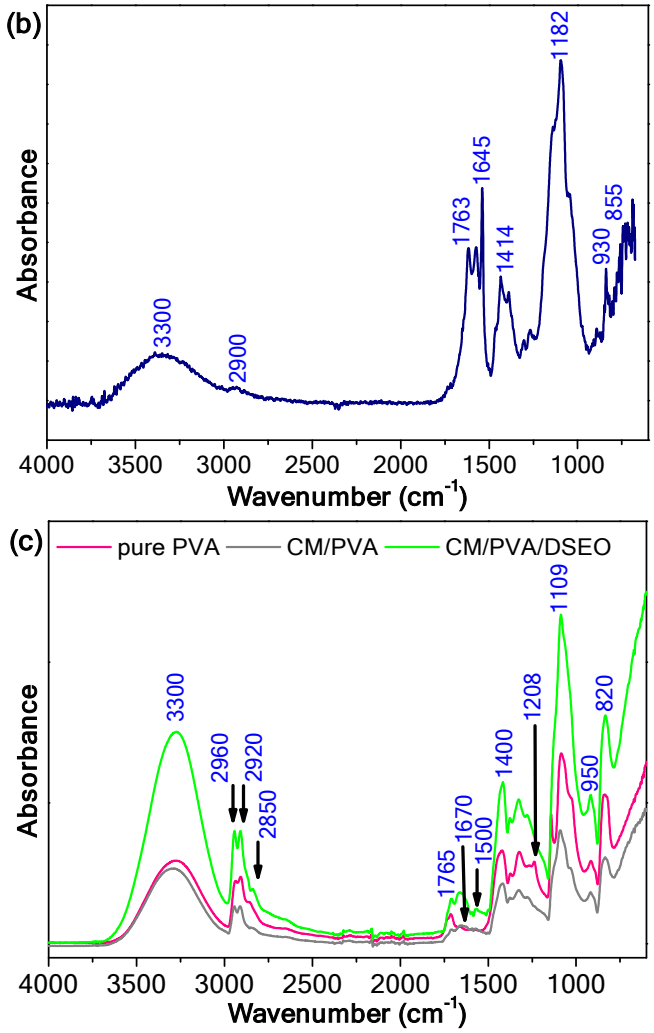


Figure 5. ATR-FTIR spectra of: (a) DSEO, (b) CM and (c) pure PVA, CM/PVA and CM/PVA/DSEO electrospun nanofibres.

The two bands observed at 1645 and 1414 cm^{-1} are attributed to the asymmetric and symmetric stretching vibrations of the $–COO^-$ of the salts of the carboxylic acids, probably the uronic acid in mucilage.[6]The characteristic band shown at 1182 cm^{-1} corresponds to the C−O stretching vibration of the pyranose ring of mucilage.[57,60] The bands observed at 930 and 855 cm^{-1} are

attributed to the presence of α and β-D-glucose configurations.[57] The studied spectra reveal that cactus mucilage is rich in polysaccharides, which can form new links with other polymers.

Figure 5c shows the chemical functional profiles for the pure PVA, CM/PVA and CM/PVA/DSEO electrospun nanofibers. The spectra of the analyzed electrospun nanofibers did not show significant changes when compared to those of pure PVA. For pure PVA, the broadband absorption at 3300 cm^{-1} corresponds to hydroxyl stretching vibrations for the intermolecular and intramolecular hydrogen bonds. The bands at 2960 and 2920 cm^{-1} are attributed to the C–H stretching vibrations of alkyl groups, and the peaks in the region between 1765 and 1700 cm^{-1} are assigned to the non-hydrolyzed vinyl acetate groups (C=O stretching).[53] Other peaks at 1400, 1109, 950, and 820 cm^{-1} are related to the CH_2, C–O–C and C–C bonds, respectively.[61] The spectra of CM/PVA electrospun nanofibers revealed increased absorption intensities at 1670 and between 1100 and 1200 cm^{-1}, corresponding to both carbonyl and carbon-oxygen stretching frequencies, respectively, associated with the polysaccharide nature of mucilage.[62] In the spectra of CM/PVA/DSEO electrospun nanofibers, the addition of DSEO did not cause any significant changes compared with the spectra of CM/PVA electrospun nanofibers. It could be seen that the DSEO and CM/PVA polymers are intermolecularly bound, which

confirms the encapsulation of oil in the CM-blend PVA nanofiber structure. These findings agree with the other studies in the literature.[17,22,50,53,62]

3.4. Thermal properties

The TGA/HF profiles of DSEO, CM, pure PVA, CM/PVA and CM/PVA/DSEO electrospun nanofibers were recorded at temperatures ranging from 30 to 700 ˚C (Figure 6). The TGA of DSEO is presented in Figure 6a. As can be seen, DSEO has a typical TGA profile with only one evaporation stage starting at 70 ˚C and finishing at 200 ˚C, with a maximum weight loss of 99.6wt% around 110 ˚C. From the HF curve, it can be seen that DSEO absorbed energy to assess the evaporation. The thermal behavior of DSEO is similar to that of the essential oil of citrus limon[50] and greater than that of cinnamon essential oil, which has an evapouration/degradation temperature between 63 ˚C and 119 ˚C.[10]

The TGA curve of CM (Figure 6b) revealed three essential degradation zones. The first zone is kept between 30 and 150 ˚C, degrades about 12 wt% of CM, and involves the evaporation of the moisture.[36,63] The second zone ranges between 150 and 490 ˚C and is attributed to the mucilage's thermal decomposition with a loss of 40 wt%. A slight thermal stability appears between 150 and 250 ˚C due to gelatinization followed by branch breakage.[36,38] When the CM is

heated, thermal decomposition occurs in this zone, involving the continuous and slow decomposition of carbonaceous matter. The HF curve of CM shows the combustion peak, which releases a high quantity of energy during the decomposition reaction. The third zone occurs from 490 to 700 ˚C, in which losses of 18 wt% follow slight thermal stability and involve broken chains, residual branches of polysaccharides, and some crystalline structure degradations.[64] Beyond 680 ˚C, the slight stability revealed the ash content.

The TGA curve of PVA displays two thermal events (Figure 6c). The first thermal decomposition zone occurs between 30 and 210 ˚C, with losses of about 8 wt%. It involves the evaporation of physically weakly bound water and chemically strongly bound water due to the relaxation of the crystalline domains in PVA.[65] The second transition zone ranges from 210 to 530 ˚C and loses about 92 wt%. The total weight loss of pure PVA was reached at 490 ˚C. This event is confirmed by the typical prominent peak observed in the HF curve (Figure 6d). This significant transition is associated with the degradation of the PVA polymeric backbone caused by the melting of the crystalline domains as hydrogen bonds begin to break.[31,65]

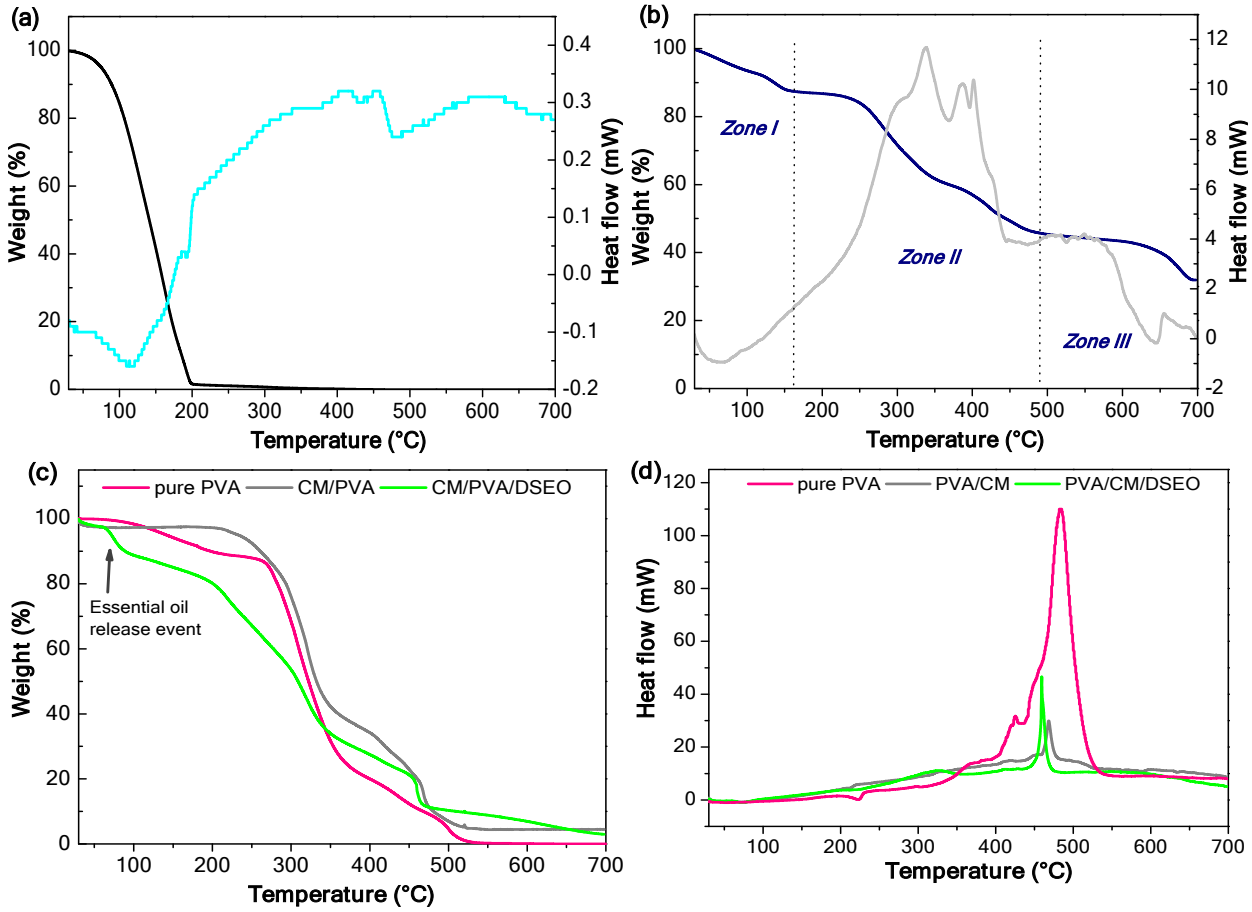


Figure 6. TGA thermograms of: (a) TGA/HF curves of DSEO, (b) TGA/HF of CM, (c) TGA and (d) HF curves of pure PVA, CM/PVA and CM/PVA/DSEO electrospun nanofibres.

The TGA curve of CM/PVA displays two thermal events (Figure 6c). The first step of degradation ranges from 30 to 120˚C; losses of about 3 wt% are attributed to the evaporation of moisture at 80 ˚C (Figure 6d). The second step starts from 120 ˚C to 530 ˚C and causes a degradation of about 92 wt%. Thermal stability occurs in the region of 120 and 230 ˚C related to the CM enhancement of PVA electrospun; similar stability is shown for CM in Figure 6b. The

second step of degradation includes two transitions. The first transitional degradation began at 250 ˚C and was achieved at 350 ˚C attributed to the heating degradation of the PVA side chain.[62] The second transition occurs from 350 to 530 ˚C and corresponds to continuous mucilage polysaccharide degradation. The higher thermal stability of CM/PVA electrospun nanofibers compared to pure PVA might be due to the hydrogen bonding interaction between the hydroxyl groups of CM and PVA and the compactness of the chain. The CM/PVA electrospun nanofibers produced in this study are more thermally stable than the basil seed mucilage/PVA electrospun nanofibers[49] and flaxseed mucilage/PVA nanofibers.[66]

The TGA curve of CM/PVA/DSEO electrospun nanofibers shows that the weight loss occurred mainly in two stages with significant changes in CM/PVA electrospun nanofibers (Figure 6c). From 30 to 120 ˚C, the evaporation of residuals has occurred, and the oil releases when heated above 70 ˚C. The second stage of degradation starts at 120 to 480˚C. In this region, two thermal transitions of degradation appeared. The first transition occurs between 120 and 350˚C attributed to PVA backbone polymer degradation, while the second transition ranges from 350 to 480˚C and corresponds to polysaccharide degradation. After 480˚C, an ultimate line of weight loss was observed to slow down, related to the presence of ash naturally present in cactus mucilage

polysaccharides. The findings suggest that adding cactus mucilage enhances the thermal stability of PVA electrospun nanofibers and that the DSEO is successfully encapsulated in the nanospun fibers. The thermal behavior of CM/PVA/DSEO electrospun nanofibers is more significant than that of PVA/cinnamon essential oil and PVA/cinnamon essential oil/β-cyclodextrin electrospun nanofibers.[50] The remarkable difference in the TGA curves of DSEO, CM, pure PVA and electrospun nanofibers revealed that the chemical interaction among the two polymers with DSEO had occurred successfully in the electrospun nanofibers.

3.5. Contact angle

Figure 7 shows how the water contact angles of CM/PVA and CM/PVA/DSEO electrospun fibers change over time. T_0 is the initial contact time of the water droplet and T_f is the time when the analysis is over. All the water contact angles were < 90, indicating the hydrophilic property of the produced CM-blend PVA electrospun nanofibers. Hydrophilic surfaces present better affinity for cells than hydrophobic surfaces.[67]

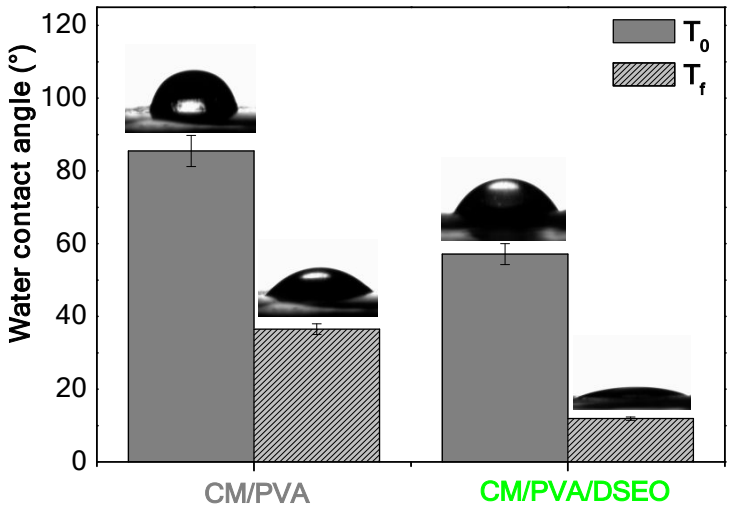


Figure 7. Water contact angles of CM/PVA and CMPVA/DSEO electrospun nanofibers.

The water contact angles of CM/PVA and CM/PVA/DSEO electrospun nanofibers were 86˚ and 58˚ at T_0, which were reduced to 37˚ and 12˚, respectively. The high water contact angle of the analyzed specimens can be explained by the rough surface of electrospun nanofibers. The recorded water contact angles of blended PVA electrospun nanofibers were observed to be greater than those of neat PVA electrospun fibers.[11] It reveals that adding cactus carbohydrates and essential oils to electrospun nanofibers can significantly reduce the value of the water contact angle. Moreover, the recorded digital pictures show clearly that the water contact angle of the electrospun nanofibers at T_0 has a better resistance against water than that of the emulsion-based electrospun nanofibers. At T_f, the reduction in contact angle values depends on the polar hydroxyl group of the formulated nanospun, especially the hydroxyl groups of cactus mucilage,

as well as the electrospun fibers partial dissolution during the water contact angle test. It means CM/PVA and CM/PVA/DSEO electrospun nanofibers have water absorbency as required for an ideal scaffold, which must be used in tissue wound dressings[11,50] and smart food packaging applications.[9]

3.6. Encapsulation efficiency

EE is one of the critical parameters in encapsulation technology; it was used to determine the efficiency of the electrospun fiber in the entrapment of the oil.[24,68] The EE of DSEO in CM/PVA electrospun nanofibers was 100%. The EE of DSEO in CM/PVA electrospun nanofibers is greater than those of thyme essential oil in zein with an EE of 75.23±16.4%,[21] rosemary ethanoic extract in PVA fibers with a retention efficiency of 88%,[69] and costmary essential oil in salep-PVA with an EE ranging between 93.21–96.62%.[22] These findings confirm the efficacy of PVA blended with cactus mucilage in the electrospinning method and for encapsulating essential oils.

3.7. Essential oil release

The release profiles of free DSEO and DSEO encapsulated in electrospun nanofibers were studied for 96 and 552 h, respectively. Figure 8 shows the release process of free DSEO which reveals that after 8 h, the release process started to increase gradually and reached 96 h to

complete the total release. The free DSEO release profile is considered more stable than that of free orange essential oil, which completes its total release after 72 h.[20]

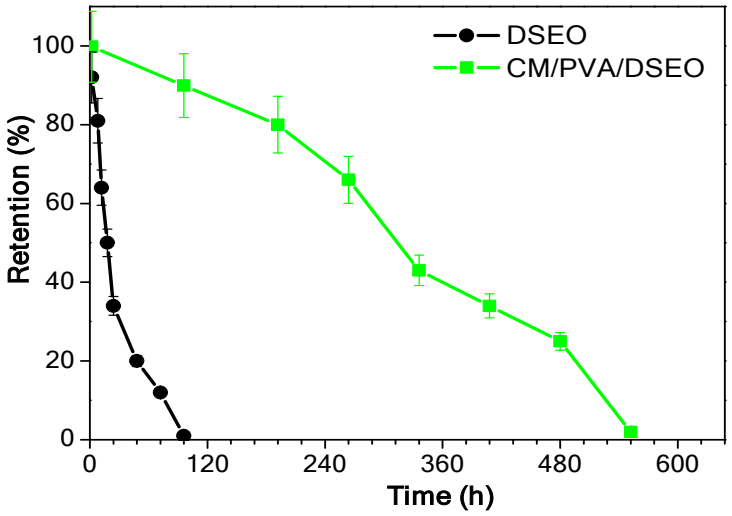


Figure 8. The release profile of free and loaded DSEO in electrospun nanofibers during 552 h (23 days).

The release process of encapsulated essential oil from electrospun nanofibers proves the gradual release process. This slower release of DSEO seems to be due to the gradual release of entrapped material, which confirms that the DSEO is encapsulated into the nanospun fibers. These findings confirm that the DSEO release took place in the fibers, suggesting that the blending of CM with PVA promotes a more controlled release. The CM/PVA electrospun fibers are considered better carriers for essential oils than the electrospun poly (lactic acid) nanofiber loaded with *Zingiber*

Cassumunar Roxb essential oil, reaching their maximum release of about 80% after 12 h[70] and almost 50% of the nanoemulsified *Cuminum cyminum* essential oil was released from the electrospun *Plantago psyllium* L. seed gum/gelatin nanofibers within the first 12 hours and reached zero after 36 h.[71] It means CM/PVA/DSEO electrospun nanofibers have a slow release process for essential oils and are suitable for innovative active packaging and pharmaceutical uses.

3.8. Anti-bacterial activity

The electrospun nanofibers were studied to assess their antibacterial activity against food-poisoning bacteria, including two strains of Gram-positive bacteria (*Staphylococcus aureus* and *Bacillus cereus*) and three strains of Gram-negative bacteria (*Pseudomonas aeruginosa, Salmonella typhi, and Escherichia coli*) using the disc diffusion method, and the obtained results were summarized in Table 3.

Table 3. Antimicrobial test of CM/PVA and CM/PVA/DSEO (10 mg/mL) against some bacterial strains of food poisoning diseases.

Bacterial strains	Inhibition zones (mm)

		CM/PVA	CM/PVA/DSEO	Gentamicin (10 µg/mL)	DMSO
Gram (−ve) PB	*Pseudomonas aeruginosa*	14.1±1.3	15.5±2.4	16.3±1.1	NI
	Salmonella typhi	13.9±2.5	12.4±1.7	16.1±1.8	NI
	Escherichia coli	12.9±2.6	15.1±0.7	15.8±0.6	NI
Gram (+ve) PB	*Staphylococcus aureus*	15.3±2.4	16.1±2.1	17.4±0.9	NI
	Bacillus cereus	14.2±1.1	14.9±3.3	15.4±1.1	NI

PB: Pathogenic Bacteria; NI: Non inhibition

Disks containing DMSO had no inhibitory effect, while gentamicin disks had an inhibitory effect on *B. cereus* and *E. coli*. The results obtained by agar disk diffusion revealed that the studied samples were potentially effective in suppressing the microbial growth of food-poisoning bacteria with varying potency. DSEO-loaded CM/PVA was the most effective nanofiber in retarding the microbial growth of all pathogenic bacteria tested at a concentration of 10 mg/mL. It appears that *E. coli* is the most resistant to CM/PVA, while *S. typhi* is the second most resistant. This is due to the nature of mucilage polysaccharides, which have good resistance against bacterial growth, unlike pure PVA, which has no inhibitory effect on bacterial growth.[72] This result is greater than that reported for nanofibers of chia seed mucilage and polyvinyl alcohol, which have no antibacterial properties.[16] However, with CM/PVA/DSEO, *S. typhi* is the

most resistant, followed by *B. cereus*. On the other hand, *S. aureus* and *P. aeruginosa* are the most sensitive strains to both types of electrospun nanofibers. CM/PVA electrospun nanofibers loaded with 4% DSEO had the highest antibacterial activity compared to that reported for electrospun *Plantago psyllium* L. seed gum/gelatin nanofibers incorporated with 3% *Cuminum cyminum* essential oil, which also has an inhibitory effect on the growth of *S. aureus*.[71] Furthermore, The improved antimicrobial activity of nanofibers loaded with essential oil is significantly due to the existence of antibacterial active compounds in DSEO, essentially d–limonene, apiol, piperitone, and carvone (Table 1). *d*–limonene is the main compound present in DSEO; it has great applications in antibacterial and food preservation due to its bactericidal activity, safety, and low toxicity.[72−74] Also, DSEO has an important antibacterial effect against foodborne pathogenic bacteria.[44]

The MIC's of the CM/PVA and CM/PVA/DSEO nanofibers were determined by disc diffusion method to evaluate their bacteriostatic and bactericidal properties, and the obtained results are reported in Table 4. The inhibitory effect of CM/PVA/DSEO started at 2.5 mg/mL with inhibition zones of 9.3 and 8.4 mm against *S. aureus* and *P. aeruginosa* while CM/PVA/DSEO nanofibers suppressed the bacterial growth of these strains at a concentration of 2.5 mg/mL with

inhibition zones of 6.8 and 7.6 mm, respectively. The MBC's were confirmed by the absence of bacterial growth in the tested strains streaked from the inhibition zone corresponding to their lowest MIC's. The CM/PVA nanofibers showed potentially bactericidal activity against *S. aureus and P. aeruginosa* with a MBC of 5 mg/mL whereas the MBC of CM/PVA/DSEO reached 10 mg/mL exceptionally for *P. aeruginosa* which is less sensitive with a MBC in the range of 12.5 mg/mL.

Table 4. MIC's of the CM/PVA and CM/PVA/DSEO electrospun nanofibers against *S. aureus* and *P. aeruginosa.*

Sample	Conc. (mg/mL)	Inhibition zones (mm)	
		Gram (+ve) PB *S. aureus*	Gram (−ve) PB *P. aeruginosa*
CM/PVA	2.5	9.3±0.97	8.4±1.20
	5	11.3±1.10	11.3±1.10
	10	15.4±2.60	14.1±1.30
	12.5	19.4±0.92	16.3±0.67
	15	19.5±2.10	19.3±0.90
CM/PVA/DSEO	2.5	6.8±1.25	7.6±1.26
	5	9.6±1.42	10.5±0.95
	10	16.1±2.10	15.5±2.40

	12.5	19.6±1.60	20.1±1.90
	15	22.6±2.20	22.4±2.60

PB: Pathogenic Bacteria

Generally, it has been reported that the MIC and MBC of *Anethum graveolens L.* varied depending upon the plant and pathogen strains as well as the solvent system of active compound extractions.[28] These results prove the good synergy between CM and DSEO only against bacteria, compared to that of DSEO, which has MIC and MBC values ranging from 0.048 to 0.097 mg/mL and 12.5 to >50 mg/mL, respectively, for the bacterial strains.[44] The studied MIC and MBC of the electrospun nanofibers suggest that they can be used to control and take necessary measures to prevent foodborne bacteria and food poisoning. Thus, the encapsulation of DSEO in electrospun nanofibers significantly proves its effectiveness to be used as a considerable natural antibacterial agent and could be exploited as a natural preservative in the food industry owing to its efficacy and safety.[44]

4. CONCLUSION

The present study investigated the potential of cactus PVA and PVA electrospun nanofibers as wall materials for the encapsulation of DSEO via electrospinning. The GC/MS analysis of the

extracted DSEO identified four major components: *d*-limonen, apiol, piperitone, and carvone. The spinning solution and emulsion prove their suitability and efficiency to produce electrospun nanofibers. The optical microscopy of the CM/PVA/DSEO emulsion shows that it contains polydispersed microcapsules of various sizes. The morphology of the electrospun nanofibers produced without and with essential oil revealed that adding essential oil leads to a slight increase in fiber diameters. Similar findings are proven with AFM. A thermal investigation revealed the enhancement of PVA's thermal stability when blended with CM. The water contact angle measurements confirmed that formulated electrospun nanofibers have water absorbency. The encapsulation efficiency and oil release of the free and encapsulated oils prove the suitability of CM and PVA blends as effective core shells for protecting essential oils or other bioactive compounds. The CM/PVA nanofibers containing DSEO showed enhanced antibacterial properties against *S. typhi* and *B. cereus*. These results confirm that these nanofibers could be used as a promising controlled delivery system for value-added smart food packaging applications, encapsulation technologies, biomedical, and pharmaceutical applications.

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. F.M.: conceptualization, investigation, data curation, writing — original draft preparation, methodology. H.E.: resources, methodology, investigation, conceptualization. A.F.: data curation, investigation. I.O.: material preparation, conceptualization. M.N.B.: methodology, formal analysis, supervision, writing— review and editing. Y.M.: methodology, supervision, visualization, writing— review and editing

Funding Sources

This research has been funded by Ministry of Higher Education, Scientific Research and Technology, Tunisia through project number PAQ-PAES/T2-PAES 2-15.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENT

The authors acknowledge Sami Halila and Laurine Buon from CNRS, CERMAV, University of Grenoble Alpes, who analyzed oil with gas chromatography/mass spectrometry.

ABBREVIATIONS

AFM, atomic force microscopy; ATR, attenuated total reflectance; *B. cereus*, *Bacillus cereus*; CM, cactus mucilage; DMSO, dimethyl sulfoxide; DSEO, dill seed essential oil; *E. coli*, *Escherichia coli*; EE, encapsulation efficiency; HF, heat flow; MBC's, minimum bactericidal concentrations; MIC's, minimum inhibitory concentrations; NI, Non inhibition; NIST, National Institute of Standards and Technology; PVA, polyvinyl alcohol; *P. aeruginosa*, *Pseudomonas aeruginosa*; PB, pathogenic bacteria; *S. aureus*, *Staphylococcus aureus*; *S. typhi*, *Salmonella typhi*; SEM, scanning electron microscopes; TGA, thermogravimetric analysis.

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