Ultrasonic-assisted synthesis, characterization, and release kinetics of Sweetgum (Liquidambar orientalis Miller) essential oil microcapsules as efficient antibacterial materials

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Article

Keywords: microencapsulation, sweetgum essential oil, complex coacervation, gelatin, gum Arabic

Posted Date: July 14th, 2023

DOI: https://doi.org/10.21203/rs.3.rs-3145938/v1

Additional Declarations: No competing interests reported.

Abstract

The development of new antibacterial materials is an essential global requirement. Sweetgum essential oil (SGEO) has potential biological activities including antifungal, antitermite, antioxidant, and antibacterial properties. Microencapsulation is one of the most promising ways to improve physical and chemical stability of essential oils, to prevent thermal degradation, volatilization, and undesirable reactions, and to control release of active components. In this study, it was aimed to synthesize the SGEO microcapsules as new antibacterial materials by ultrasonic-assisted complex coacervation of gelatin and gum Arabic. The coacervation between gelatin and gum Arabic as functions of gelatin:gum Arabic ratio, core material load, and ultrasonication time was investigated. The SGEO microcapsules were characterized by FESEM, FTIR, TGA, and XRD. The SGEO release of the microcapsules was investigated, and best fit was observed with the Korsmeyer-Peppas model. The antibacterial activity of the microcapsules against Gram–positive and Gram–negative microorganisms (*Staphylococcus aureus* and *Escherichia coli*) were also evaluated, and it was observed that the microcapsules had strong antibacterial activity against the two common pathogenic bacteria. All the results show that the SGEO microcapsules can be candidates as novel antibacterial materials and may be used to make contributions to antibacterial properties of different industrial products.

Introduction

Essential oils are volatile lipophilic compounds extracted from plants and used in foods, cosmetics, and textile products because of their chemical, functional, antimicrobial, and antioxidant properties. However, most components of essential oils are degraded by exposure to light, oxygen, high temperatures, and relative humidity that can lead to volatilization or undesirable reactions such as oxidation. Microencapsulation technology is one of the most promising ways to protect active agents from oxidation, to prevent evaporation of volatile compounds, and to control the release of oil components [1].

Microencapsulation is a process where any active material called core material is surrounded and enclosed by natural or synthetic materials called wall materials to form microcapsules. Several methods for microencapsulation have been used, including ionic gelation [2], liposomal entrapment [3], extrusion [4], spray chilling/cooling [5], complex coacervation [6], and interfacial polymerization [7]. Complex coacervation is the favored method due to its higher stability and lower cost compared to other techniques [8]. Complex conservation also has many advantages, such as high core material loading, good controlled-release, and high encapsulation efficiency [9].

Sweetgum essential oil (SGEO) is a resinous exudate harvested from sweetgum trees (*Liquidambar Orientalis* L.) belonging to the Hamamelidaceae family, which is an endemic species that grows in Mugla, Turkey [10]. It is known as a traditional medicine in Turkey and has also been used for the treatment of some skin diseases [11] and peptic ulcers [12]. In literature studies, sweetgum oil was also reported to exhibit several biological effects including antifungal [11], anti-ulcerogenic [13], antitermite [14], antioxidant [15], nematicidal [16], antibacterial [17] activities as well as human health protection from

cytotoxic, genotoxic, metabolic actions of environmental toxicants [18] and inhibitory effects towards pancreatic lipase to manage overweight and obesity [19]. Its high antioxidant and antimicrobial properties provide the use of sweetgum in a broad range of potential applications, including in food products [20], wound dressings [21], tissue engineering [22], cosmetics [23], and pharmaceuticals [12].

To date, sweetgum essential oil has been examined in a limited number of studies in the literature, although it has superior biological activities. To the best of our knowledge, the microencapsulation of sweetgum essential oil has not been reported yet. However, microencapsulation of sweetgum essential oil is of great importance in terms of protecting its active agents against environmental conditions, maintaining its effectiveness for a long time and controlling the release rate depending on the intended usage. In this study, it was aimed to synthesize the sweetgum essential oil microcapsules using gelatin and gum Arabic as oppositely charged biopolymer pairs by ultrasonic-assisted complex coacervation technique and to evaluate the influence of process variables (wall material ratio, ultrasonication time and core material load) on the encapsulation efficiency of microcapsules. In addition, the chemical structure, thermal and surface characteristics, and the SGEO release behavior of the SGEO microcapsules were examined in detail. Antibacterial activity of the SGEO microcapsules against common Gram–positive and Gram–negative pathogenic microorganisms (*Staphylococcus aureus* and *Escherichia coli*) was also investigated.

Materials and Methods

Materials. SGEO, which was extracted from Anatolian Sweetgum trees, was purchased from Koycegiz (Mugla, Turkey). The oil includes 65.58% of cinnamyl cinnamate, 19.89% m-methyl styrene, 4.57% of cinnamic acid, 2.79% of acetocinnamon, 2.47% 1-phenyl-3-trimethylsilyloxy, 0.84% 3-Phenyl-1-propanol, 0.56% of 4-hydroxycinnamyl alcohol. Gum Arabic (9000-01-5, E414) and gelatin (AG060221, E441, 220 Bloom, average molecular weight of 45,000–50,000 Da) from bovine were purchased from Alfasol (Istanbul, Turkey). Glutaraldehyde (50 wt. % solution in ethanol) was provided by Acros Organics. n-Hexane (analytical reagent), isopropyl alcohol, ethanol (99.9%), hydrochloric acid (HCl, 37%), and sodium hydroxide (NaOH, reagent grade) were obtained from Merck Company (Germany). All chemicals used in this study were of reagent grade and used without any further purification. All aqueous solutions were prepared with deionized water.

Microencapsulation of SGEO. The SGEO microcapsules were prepared by complex coacervation via a glutaraldehyde cross-linking method [24]. The best complex coacervation condition was investigated from experiments conducted with different wall material ratio, ultrasonication time, and core material load shown in Table 1.

Table 1
Gelatin:gum Arabic ratio, ultrasonication time, and core material load tested for the microencapsulation process.

Sample	Gelatin:gum Arabic ratio	Ultrasonication time (minutes)	Core material load (g)
12G/GA-5SGEO-5t	12:1	5	5
10G/GA-5SGEO-5t	10:1	5	5
8G/GA-5SGEO-5t	8:1	5	5
6G/GA-5SGEO – 5t	6:1	5	5
6G/GA-5SGEO-10t	6:1	10	5
6G/GA-5SGEO-15t	6:1	15	5
6G/GA-5SGEO-20t	6:1	20	5
6G/GA-3.5SGEO-20t	6:1	20	3.5
6G/GA-1.75SGEO-20t	6:1	20	1.75

Briefly, aqueous solutions of gelatin (6%, 5%, 4%, and 3% w/w) were prepared separately by heating under constant stirring at 40°C in a jacketed glass reactor. The sweetgum oil (1.75, 3.5, and 5 g) was added to the gelatin solution and emulsified in an ultrasonic homogenizer ultrasonicator device (Bandelin, HD4100, Germany) at 20 kHz, 50% amplitude and no pulsation ultrasound regime for determined times (5, 10, 15, and 20 minutes). Then, 50 mL of gum Arabic solution (2.5%, w/w) were added to the emulsion and stirred by a magnetic stirrer (Isolab Laborgerate GmbH, Germany) for another 30 min. Then, the pH was reduced to 4.0 by dropwise adding 0.5 N hydrochloric acid solution and the mixture was stirred for 25 min to form a stable emulsion. The temperature of the emulsion was gradually reduced to 10°C in an ice bath during coacervation. For cross-linking of the microcapsules obtained by coacervation, 10 mL of glutaraldehyde solution (50%, w/v) was slowly added while stirring at 1000 rpm in an ice bath. After 1 hour, the obtained microcapsules were separated by centrifugation (Nuve, NF 400, Turkey) for 15 min at 4000 rpm and 25°C, and dried at 30°C in an oven (Natural Convection Oven, JSON-100, Republic of Korea).

The SGEO microcapsules synthesized with different wall material ratios, ultrasonication time, and core material load were referred to as *x*G/GA-*y*SGEO-*z*t, where *x* and *y* denote the amount of gelatin (g) and SGEO (g), respectively, and *z* denotes the ultrasonication time (minutes) applied in the synthesis process.

Determination of surface oil, total oil, and encapsulation efficiency. Both the surface oil, also known as the non-encapsulated oil fraction, and total oil contents of SGEO microcapsules were determined according to Böger et al. with modifications [25].

For surface oil analysis, about 1.5 g of dried microcapsules were added to hexane (15 mL), and the mixture was stirred manually for 2 min at room temperature without microcapsule destruction, followed by suspension filtration through a Buchner funnel. After filtration, the microcapsules were washed three times with 20 mL of hexane through the filter paper and then the solvent was evaporated from the glass bottle. The extracted oil, which was considered as the surface oil, was determined gravimetrically. Surface oil content (SO, %) was determined by the ratio of surface oil mass to dry microparticle mass [26].

In order to determine the total oil content of SGEO microcapsules, approximately 0.5 g of microcapsules were dispersed in 4 mL of hydrochloric acid (4 N) in a sealed container and agitated for 10 min at 40°C. Subsequently, 15 mL of a solvent mixture (hexane:isopropanol, 3:1, v:v) was added into this mixture and stirred in an automatic shaker at 100 rpm to speed up the extraction of the oil into the solvent phase. This dispersion was then centrifuged at 4,000 rpm for 10 min and the upper phase containing the dissolved oil was then separated in a preweighed beaker. The extraction step was repeated as described above and all supernatants collected in the beaker were evaporated at 80°C. Finally, the recovered oil was oven dried at 50°C until constant weight. The oil content was determined gravimetrically and used to calculate the total oil content (TO %) as the ratio of the extracted oil mass to the dry microparticle mass. Measurement of the total oil and surface oil was carried out in triplicate for each sample.

Encapsulation efficiency was calculated based on the oil present on the surface of the microcapsules and the total oil present in the microcapsules. The percent of encapsulation efficiency was calculated using Eq. 1 [27]:

$$EncapsulationEfficiency\% = \frac{W_{TO} - W_{SO}}{W_{TO}} \times 100$$

1

where W_{TO} is the weight of total SGEO in microcapsules and W_{SO} is the weight of surface oil.

Characterization. The chemical structure of gum Arabic, gelatin, SGEO, and SGEO microcapsules with the highest encapsulation efficiency was characterized using a Fourier Transform Infrared Spectrometer (FT-IR, Thermo Scientific Nicolet iS50FT-IR). The spectral data of all samples were acquired in the wavenumber range of 400–4000 cm^{-1} with 50 scans at a resolution of 4 cm^{-1}. The thermal behaviour of the samples was investigated via thermogravimetric analysis (TGA). A quantity of each type of sample was placed in a sealed aluminium pan and analysed by Simultaneous Thermal Analyzer (STA, Netzsch STA 449 F3, USA) operating at a heating rate of 10°C/min in a dynamic nitrogen atmosphere (50 mL/min). The TGA thermograms of the samples were recorded using NETZSCH Proteus Thermal Analysis software. The X-ray diffractograms of the samples were obtained using an X-ray diffractometer (XRD-6000, Shimadzu, Japan) with CuKα line radiation (λ = 1.544 Å). The samples were loaded on a sample holder and scanned over the angle (2θ) range of 10–80° with the step size of 0.02°/min. The morphology of the SGEO microcapsules was investigated with a Field Emission Scanning Electron

Microscopy (FESEM, Gemini Supra 40 VP, Zeiss, Germany) at an accelerating voltage of 20 kV. The microcapsules were coated with a thin layer of gold using a sputter coater (Quorum Q150R ES, United Kingdom). The magnification of SEM was recorded at 1000× and 3000×.

Release of SGEO from microcapsules. The release rate of SGEO from the microcapsules was determined as a function of time using a UV–VIS spectrophotometer. A known amount of SGEO microcapsules was added to a mixing and temperature-controlled vessel containing 20 mL of ethanol at 25°C. At regular time intervals, samples (3 mL) were taken from the suspension, which was continuously stirred at around 300 rpm. The samples were analysed by UV-VIS spectroscopy (Optizen UV/Vis spectrophotometer, Mecasys Co. Ltd., South Korea) by measuring the absorbance at 275 nm, which was the maximum absorbance of SGEO in ethanol. The SGEO released concentration was calculated using a previously obtained calibration curve of SGEO in ethanol. Concentrations from 0.1 to 1.0 mg/mL were prepared. A 1:50 dilution factor was used to read the absorbance. The cumulative release of SGEO was calculated from the mass of SGEO released in time *t* and the initial mass of SGEO loaded in the sample. All experiments were carried out in triplicate and reported as average values plotted with standard deviation errors.

Determination of antibacterial activity. Antimicrobial activity of the SGEO microcapsules was evaluated following ASTM E2149-13a [28]. 1±0.1 g of sample was placed in 250 ml Erlenmeyer flasks and covered with 100 ± 0.1 ml of the bacteria-containing buffer. A bacteria suspension without samples was used as reference. Flasks were placed in a shaker thermostated at 100 rpm. Incubation was performed for 24 h, at 35±1. To prepare the bacterial inoculum, *Escherichia coli* ATCC 25922 and *Staphylococcus aureus* ATCC 6538 bacteria were transferred to sterile Tryptic Soy Broth medium and cultivated for 18 h at 35±2 °C. Then, the culture was diluted with the sterile buffer solution to obtain a final concentration of 1.5 × 105 CFU/mL. Decimal dilution of the bacteria suspension was made, followed by the pour plate method, using three parallel plates for each of the dilutions. After 24 hours culture at 37°C, colonies were counted and Reduction % (CFU/mL) was calculated using Eq. 3:

$$Reduction\backslash\%\,(CFU/mL) = \left(\frac{B-A}{B}\right) x100$$

3

where A and B are CFU/mL for the flask containing the treated substrate after the specified contact time and CFU/mL for the “inoculum only” flask after the specified contact time, respectively.

Results and Discussion

Effect of process parameters on encapsulation efficiency. Successful microencapsulation process of essential oils results in a microcapsule with both minimum surface oil content and maximum retention of the core material inside the microcapsules. In this study, the effect of wall material ratio, ultrasonication time, and core material load on the encapsulation efficiency of SGEO through the

complex coacervation process was investigated. Table 2 exhibits the different microcapsule formulations and the result of encapsulation efficiency values of SGEO. Encapsulation efficiency values of the SGEO microcapsules synthesized in this study were obtained in the range of 80.06–95.28%. The conditions that provided the highest encapsulation efficiency were determined as follows: the ratio of gelatin:gum Arabic of 6:1, the core material load of 5 g, and ultrasonication time of 20 min, at which conditions encapsulation of the SGEO was carried out effectually.

Table 2
Surface oil, total oil, and encapsulation efficiency of SGEO microcapsules.

Sample	Total Oil (%)	Surface Oil (%)	Encapsulation Efficiency (%)
12G/GA-5SGEO-5t	11.71±0.46	2.33±0.10	80.06±1.64
10G/GA-5SGEO-5t	12.21±0.27	2.22±0.14	81.81±1.46
8G/GA-5SGEO-5t	13.13±0.21	2.07±0.05	84.21±0.17
6G/GA-5SGEO – 5t	13.97±0.08	1.48±0.15	89.43±1.01
6G/GA-5SGEO-10t	14.13±0.17	1.29±0.03	90.89±0.33
6G/GA-5SGEO-15t	16.20±0.52	1.16±0.06	92.83±0.61
6G/GA-5SGEO-20t	19.60±0.54	0.92±0.06	95.28±0.43
6G/GA-3.5SGEO-20t	14.97±0.47	0.77±0.04	94.85±0.33
6G/GA-1.75SGEO-20t	11.15±0.18	0.58±0.04	94.76±0.39
Data represented as Mean ± SD, n = 3.			

As shown in Table 2, the decreasing of gelatin:gum Arabic ratio had a positive effect on the encapsulation of SGEO. In the coating process, gelatin and gum Arabic were acted as a positive polyelectrolyte and a negative polyelectrolyte, respectively. The ratio of gelatin:gum Arabic affected the charge balance, causing a change in the intensity of oppositely charged ion interactions during complexation [29]. High amounts of wall materials in complex solution may cause an excess of charge, which that the formation of the complexes affected negatively.

As the ultrasonication time also increased from 5 to 20 min, the encapsulation efficiency of the microcapsules increased from 89.43–95.28%, which indicates that increasing ultrasonication time between 5 and 20 min had a positive effect on the encapsulation of SGEO. It was also observed that the shorter ultrasonic treatment time than 5 min was not sufficient to prepare a homogeneous emulsion, and the higher ultrasonic time than 20 min increased the emulsion temperature because of the high cavitation effect. The emulsion temperature may cause a decrease in the embedding rate of essential oil in polymeric materials due to the essential oil volatilization [30].

When evaluating the effect of core material load on the encapsulation efficiency, it was observed that increasing SGEO load used in the synthesis process from 1.75 g to 5.00 g positively affected the encapsulation efficiency. However, the higher oil load than 5.00 g had a negative effect on the emulsion, causing the oil droplets to not show a homogeneous mixture during ultrasonic treatment. The highest encapsulation efficiency value was recorded as 95.28% for a core material load of 5.00 g.

The optimum sample was considered as 6G/GA-5SGEO-20t microcapsules with the highest encapsulation efficiency, and the 6G/GA-5SGEO-20t microcapsules were used in the thermal, chemical, and morphological analyses in the following sections.

FTIR. The chemical structure of gum Arabic, gelatin, SGEO, and SGEO microcapsules was investigated by FTIR spectroscopy analysis and the FTIR spectra are shown in Fig. 1.

Amide groups of gelatin peptide bonds exhibit characteristic absorption spectral peaks in specific bands, which are amide I (1600–1800 cm^{-1}), II (1470–1570 cm^{-1}), III (1250–1350 cm^{-1}), and A (3300–3500 cm^{-1}) bands [31]. As shown in Fig. 1, the characteristic absorption bands of the gelatin spectrum were observed at 3289 cm^{-1}, 1624 cm^{-1}, 1540 cm^{-1}, and 1238 cm^{-1}, correspond to amide A (the vibrations of OH and NH groups), amide I (the elongation vibrations of C = O and CN groups), amide II (the vibrations of NH and CN groups) and amide III (elongation of the vibrations of NH and CN groups), respectively [32]. The FTIR spectrum of gum Arabic showed a band at 3347 cm^{-1} due to the presence of hydroxyl groups. A peak that was observed with low intensity at about 2932 cm^{-1} corresponds to free carboxylic groups [33]. The carboxylic groups of gum Arabic are responsible for negative charges to promote the process of coacervation [34]. In the spectrum of SGEO microcapsules, the hydroxyl and carboxyl groups shifted from 3347 cm^{-1} to 3300 cm^{-1} and from 2932 cm^{-1} to 2943 cm^{-1}, respectively. These changes indicate that there were hydrogen bonding interactions in the formation of the microcapsules [32]. The other characteristic absorption bands of gum Arabic spectrum were observed at 1598 cm^{-1}, 1364 cm^{-1}, and 1004 cm^{-1}, related to C = O stretching/N-H bending, C-N stretching, and C-O stretching, respectively [35].

In the FTIR spectrum of SGEO, significant vibrational bands at 3420, 2943, 1706, 1161, 965, 767, and 683 cm^{-1} were observed. The weak spectral band at 3420 cm^{-1} corresponds to the stretching vibrations of the OH functional group of alcohols [36]. The band with a peak point at around 2943 cm^{-1} could be attributed to C = C–C ring vibrations of volatile compounds and it also refers to C-H vibrations of the aromatic methoxyl, methyl and methylene groups of the side chains [37, 38]. The strong and sharp absorption peaks at 1706 cm^{-1} is assigned to the C = O stretching vibration peak of ketones. The peaks observed at between 1600 and 1450 cm^{-1} were assigned to the vibrations of C = C bonds in the aromatic compounds, while the peak at between 1000 and 650 cm^{-1} were related to the deformation vibrations of C–H bonds in the benzene ring [39], the stretching vibration of C–O bond in the primary and secondary alcoholic groups [37]. The characteristic bands of SGEO were also identified in the FTIR spectra of the SGEO microcapsules. This indicates that the SGEO was successfully encapsulated into the gelatin/gum

Arabic structure. The peak at around 2850 cm^{-1} which was related to the formation of ionic interaction between the negative carboxyl group of gum Arabic and the protonated amine group of gelatin [40] were detected in FTIR spectrums of the SGEO microcapsules, while they were not visible in the gelatin and gum Arabic spectrums. The successful encapsulation of SGEO with gelatin and gum Arabic polymer shells was supported by the FTIR spectrum of sweetgum.

Thermal properties. The thermal stability of gelatin, gum Arabic, SGEO, and SGEO microcapsules was investigated by TGA, and the differential thermogravimetric (DTG) and TGA curves of the samples are depicted in Fig. 2.

As shown in Fig. 2, SGEO presents weight loss in two stages, while gelatin, gum Arabic, and SGEO microcapsules present weight loss in three stages. In the TGA thermogram of gelatin and gum Arabic, it was observed that these two wall materials present similar decomposition behavior. Firstly, a small weight loss was detected up to a temperature of about 100°C related to the departure of adsorbed and bound water. The main stage of thermal degradation was observed in the temperature range of 230 to 450°C with a weight loss of about 60.0% for gelatin because of the thermal degradation of the peptide bonds of the gelatin molecules [41] and 59.9% for gum Arabic attributed to the degradation of polysaccharides [42].

The thermogram of SGEO presents fast thermal degradation sub-divided into two steps. While the first stage observed in the temperature range of 100 to 315°C was associated with the evaporation of the simplest volatile compounds due to their heat-sensitive structure [43], the other stage observed in the temperature range of 315 to 415.5°C corresponds to the degradation of the complex aromatic rings present in the major compounds of SGEO [44]. SGEO also completely decomposes at 415.5°C, which indicates the thermal instability of its components.

In the thermograms of the SGEO microcapsules, it was observed that the SGEO microcapsules present three different stages. The first stage was observed at temperatures up to 230°C due to the loss of adsorbed and bound water present in the samples, the volatile compounds of free SGEO on the surface of microcapsules and SGEO diffused into the pores of the microcapsules, which recorded weight losses of 19.01%. The second stage was from around 230 to 450°C with average weight loss of 64.18%. The significant mass loss is associated with the depolymerization and degradation of gelatin and gum Arabic and as a result, encapsulated SGEO was released and evaporated. The third stage was observed between 450 and 600°C because of the carbonization of all the structures of microcapsules and which recorded weight loss of 7.06%. At 600°C, the residual masses are 9.75%. These results demonstrated that the thermal resistance of the SGEO microcapsules was significantly higher than un-encapsulated SGEO and the interaction between gelatin and gum Arabic in complex coacervates was enhanced thermal stability of SGEO as a heat-sensitive compound.

XRD. In order to investigate the crystallinity of wall materials, SGEO, and the SGEO microcapsules, the X-ray diffraction spectra were recorded. From the XRD patterns (Fig. 3), gelatin and gum Arabic show a prominent broad peak at around 15–25°, indicating the amorphous nature of this biopolymer with very

low crystallization [45]. The width of the XRD peak is closely related to the crystallite of the samples, and the broadened peak is usually due to the imperfect crystal structure of the samples [46]. Gelatin and gum Arabic present the characteristic broad peak at 2θ = 20.7° and 2θ = 18.7°, respectively. The microcapsules have an amorphous dispersion peak with greater intensity at about 2θ = 19.7°, which signifies the characteristic of SGEO microcapsules. The XRD spectra of SGEO microcapsules suggested that there are some changes in the original crystallization behavior of wall materials and shifting of the diffraction peak, which could be a result of emerging amide bonds between gelatin and gum Arabic [47] and hydrogen bond forces between the core material and the wall materials [30]. The XRD pattern of unencapsulated SGEO and microcapsules revealed higher crystallinity than gum Arabic and gelatin. The peak intensity of the microcapsules was higher compared to the wall materials. This phenomenon may be due to the increment in the crystallinity of the microcapsules and the homogeneous distribution of the essential oil in the biopolymer matrix [48]. SGEO exhibited a broad peak at 2θ ≈ 18.7° which could be due to organic materials adsorbed on SGEO [47]. Furthermore, the semicrystalline pattern of SGEO disappeared in the microcapsules, which confirmed the successful loading of SGEO into the microcapsules.

Morphological analysis. Figure 4 shows the SEM images of the SGEO microcapsules. As shown in Fig. 4, the SGEO microcapsules have a spherical shape with an irregular surface. The size of most microcapsules varies from about 1 μm to 20 μm. However, the microcapsules with solid walls have some cracks and small pits on their surface, which can be attributed to the drying process [40]. Elimination of water droplets on the surface of the microcapsules by oven drying may create openness in microcapsules.

Release studies. The in vitro release behavior of SGEO from the microcapsules was examined and shown in Fig. 5. The release of SGEO from the microcapsules can be divided into two phases. During the first 3 hours, there was an initial rapid release that corresponded to 72.52%, followed by a slower release up to 12 hours of 91.49% of the encapsulated SGEO amount from the microcapsules. The initial release of SGEO from the microcapsules may be relatively resulted from the part of the uncapsulated SGEO on the surface of the microcapsules, rather than the part of SGEO capsulated in the microcapsule interior.

To evaluate the mechanism of SGEO release from the microcapsules, the release data were fitted by mathematical models describing zero-order, first-order [49], Korsmeyer-Peppas [50], Higuchi [51], and Baker-Lonsdale [52] kinetic models, and the kinetic model equations are given in Table 3. In the kinetic model equations, Q and Q_0 are the cumulative amount of SGEO released at time t and the initial amount of SGEO ($t = 0$). k_0 and k_1 are the zero-order and first-order release constants. M_t/M_∞ is the percentage of SGEO released at time t relative to the percentage released at infinite time. n is a diffusion exponent indicating the type of release mechanism as Fickian diffusion (case I transport) ($n \leq 0.43$), non-Fickian or anomalous transport ($0.43 \leq n < 0.85$), or case II transport ($n \geq 0.85$). k_H is the Higuchi constant of dissolution and k_K is the Korsmeyer-Peppas model constant, which reveals structural and geometric character of the drug release matrix.

Table 3
Parameters of the kinetic models for the analysis of the release of the sweetgum oil from the microcapsules.

Kinetic Models	Equations		
Zero-order	$Q = Q_0 - k_0 t$	R^2	0.8633
		Q_0	49.0170
		k_0	6.8256
First-order	$\ln(100 - Q) = \ln Q_0 - k_1 t$	R^2	0.9891
		Q_0	57.3172
		k_1	0.2939
Korsmeyer-Peppas Model	$\frac{M_t}{M_\infty} = k_K t^n$	R^2	0.9938
		k_K	52.9792
		n	0.2275
Higuchi Model	$\frac{M_t}{M_\infty} = k_H t^{0.5}$	R^2	0.9507
		k_H	18.999
Baker-Lonsdale Model	$f = \frac{3}{2}\left[1 - \left(1 - \frac{M_t}{M_\infty}\right)^{\frac{2}{3}}\right] - \frac{M_t}{M_\infty} = K.t$	R^2	0.9342
		K	0.0233

SGEO release kinetic parameters of the microcapsules obtained by zero-order, first-order, Korsmeyer-Peppas, Higuchi, and Baker-Lonsdale kinetic models are given in Table 3. The models with higher correlation coefficients (R^2) have been considered as a more appropriate model for the evaluation of the release data. Based on R^2 as shown in Table 3, first-order (R^2 = 0.9891), Higuchi (R^2 = 0.9507), and Korsmeyer-Peppas (R^2 = 0.9938) models can be chosen as an appropriate for the explaining release kinetic of SGEO. However, Korsmeyer-Peppas is the best appropriate model with the highest R^2 value (R^2 > 0.99) for the SGEO release from the SGEO microcapsules. The diffusion exponent in the Korsmeyer–Peppas model was found to be less than 0.43. The results suggest that the Fickian behaviour of the diffusion-controlled release process is due to the low degree of swelling and the presence of oil droplets on the surface of the microcapsules [53].

Antibacterial properties. The SGEO microcapsules were investigated for their antibacterial activity against common Gram-positive and Gram-negative pathogenic microorganisms including *Staphylococcus aureus* and *Escherichia coli* according to the standard ASTM E2149-13a [28]. The antibacterial activities were evaluated after a specific contact time and calculated by reduction in the percent of *E. coli* and *S. aureus* bacteria. As shown in Table 4, it can be noted that SGEO microcapsules are effective antibacterial materials against the two common pathogenic bacteria.

Table 4
Reduction of *Escherichia coli* and *Staphylococcus aureus* exposed to the samples after 24 h.

Sample	*S. aureus* Reduction %	*E. coli* Reduction %
SGEO microcapsules	99.98	99.99

Conclusions

In this study, the microencapsulation of SGEO with gelatin and gum Arabic as wall materials depending on the wall material ratio, ultrasonication time, and core material load, which strongly influenced the encapsulation efficiency, were examined. The synthesis conditions that provided the highest encapsulation efficiency (95.28±0.43%) were determined as follows: 6:1 of the ratio of gelatin:gum Arabic, 5.00 g of the core material load, and 20 min of ultrasonication time. The characterization of the SGEO microcapsules was carried out by FESEM, FTIR, TGA, XRD and the results confirmed that SGEO was successfully encapsulated with gelatin and gum Arabic via a ultrasonic-assisted complex coacervation method. The kinetics and mechanism of SGEO release from the SGEO microcapsules were studied and the best fit was observed with the Korsmeyer-Peppas model. In addition, it was shown that the SGEO microencapsulation had strong antimicrobial activity against *E. coli* and *S. aureus* bacteria. Therefore, the successfully prepared SGEO microcapsules might have the potential for applications in the textile, cosmetic, medical, paper, food, and plastic industries as antibacterial materials.

Declarations

Acknowledgements

The MSc. study was supported by Scientific Research Projects of Pamukkale University. The authors would like to acknowledge the funding support of the Pamukkale University BAP project [ID: 2021FEBE074].

Author contributions

This study was produced from the MSc. thesis of SC completed under supervision of DAS. SC and DAS manufactured the samples, performed the measurements, processed the experimental data, performed

the analysis. DAS also was involved in planning and supervised the work, drafted the manuscript, aided in interpreting the results and worked on the manuscript.

Competing interests

The authors declare no competing interests.

Data availability

The data used to support the findings of this study are available from the corresponding author upon request.

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Figures

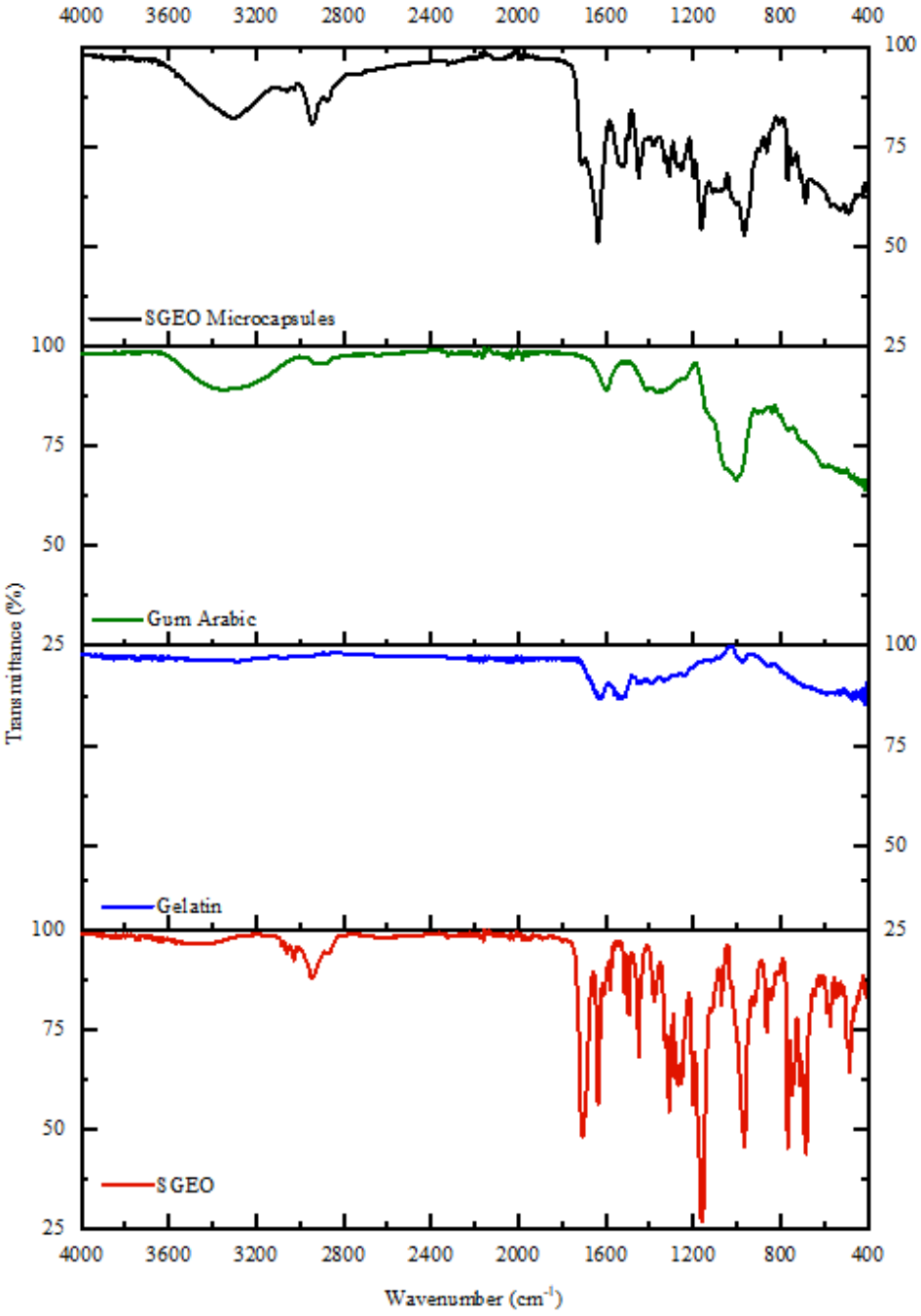

Figure 1

FTIR spectrum of gelatin, gum Arabic, SGEO, and SGEO microcapsules.

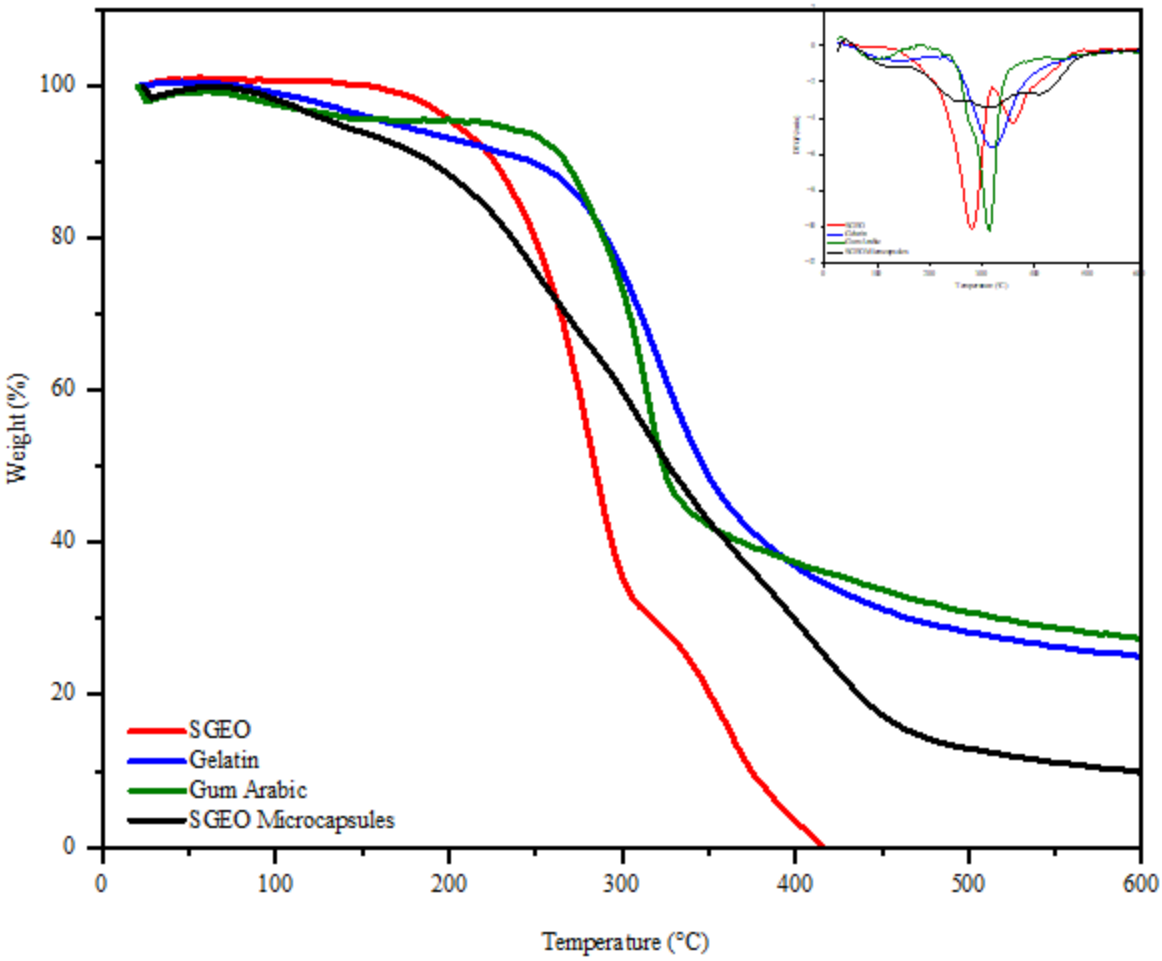

Figure 2

TGA and DTG curves of gelatin, gum Arabic, SGEO, and SGEO microcapsules.

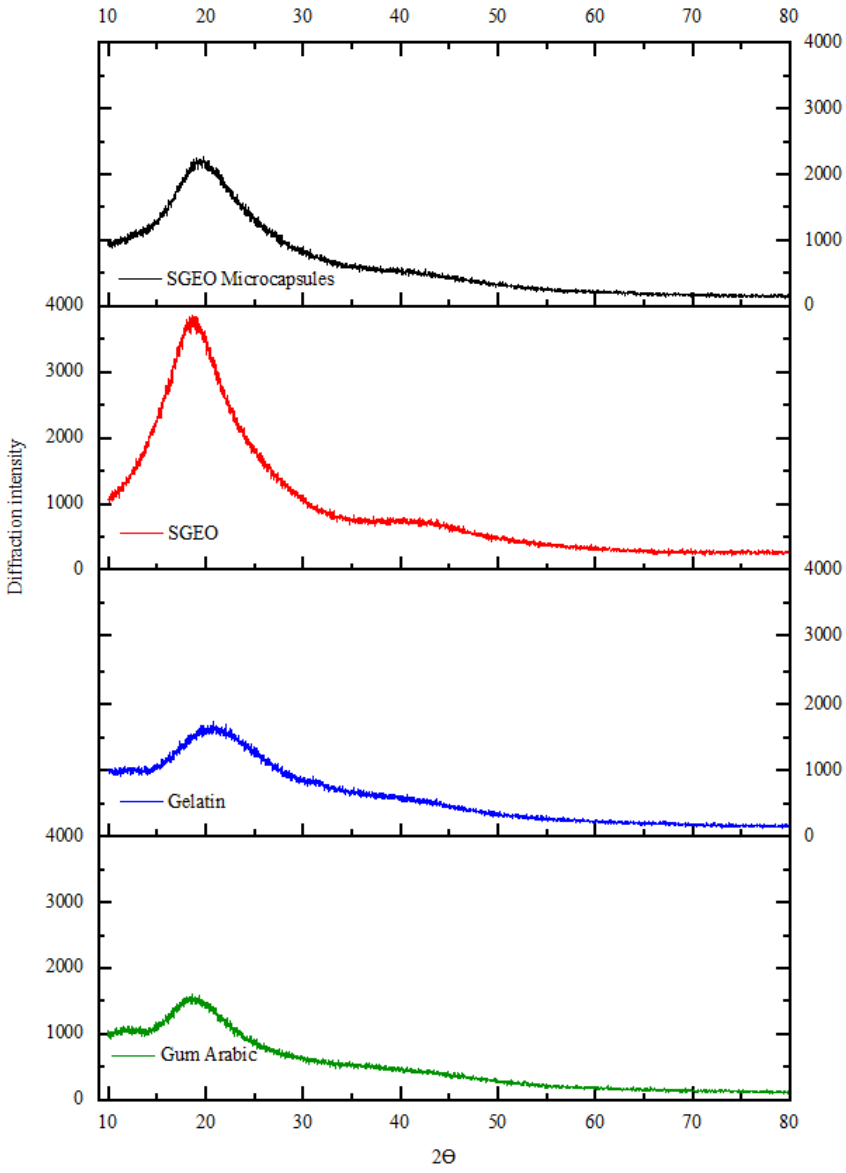

Figure 3

XRD patterns of gum Arabic, gelatin, SGEO, and SGEO microcapsules.

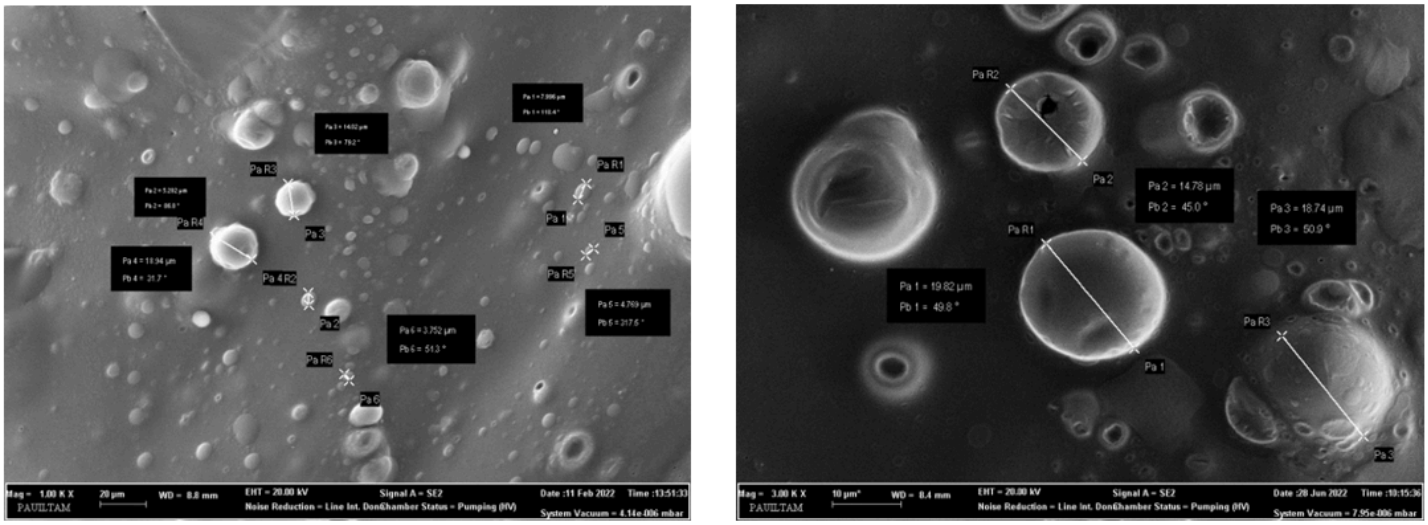

Figure 4

SEM images of the SGEO microcapsules.

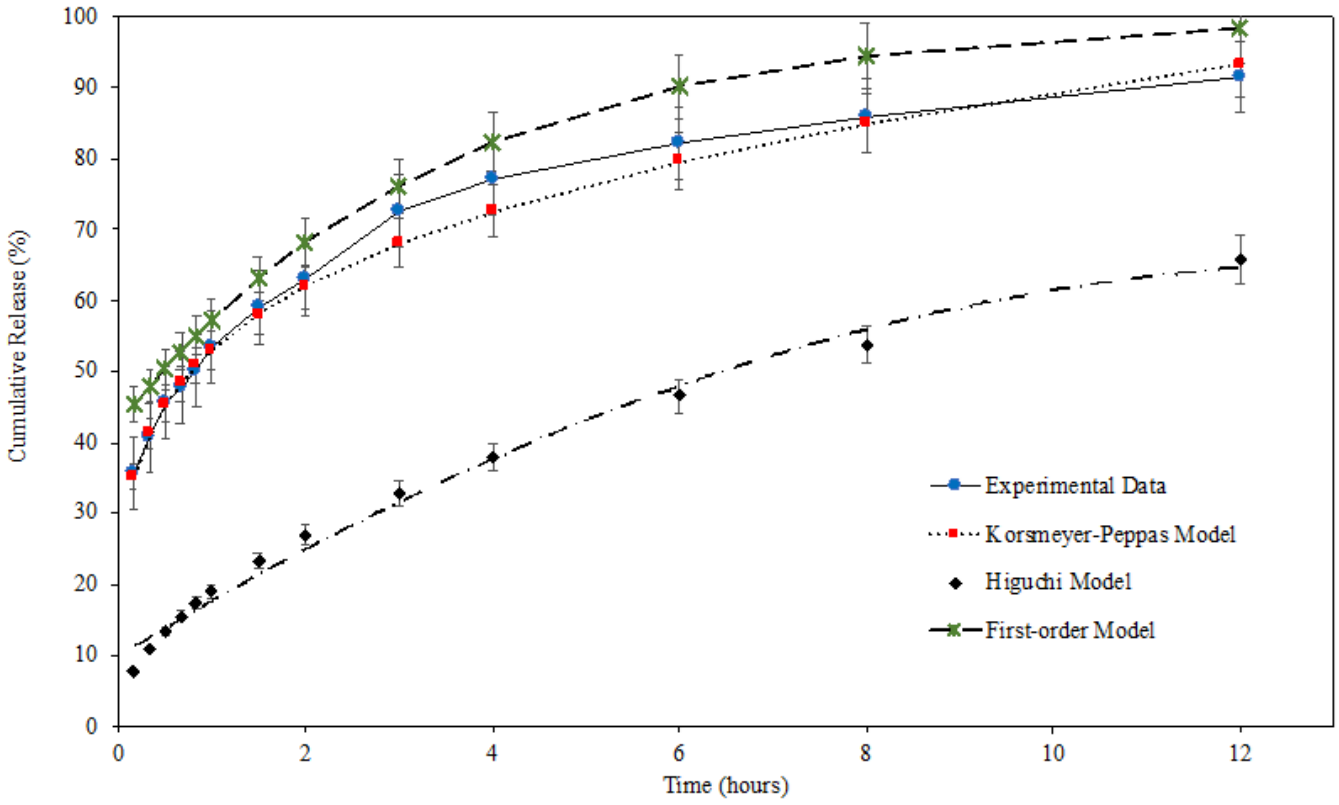

Figure 5

The release profiles of SGEO from the SGEO microcapsules. Data were represented as the mean ± SD from three experiments