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The use of mucilage extracted from *Opuntia ficus indica* as a microencapsulating shell

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Abstract: This study was aimed at investigating the micro-formulation of capsules using natural biopolymers, such as cactus mucilage (CM), carboxymethyl cellulose sodium salt (CMCNa) and chitosan (Chi) as the wall material, for the transport and supply of sunflower oil. CM samples were extracted from *Opuntia ficus indica* (OFI) by precipitation at different supernatant pH values (2, 4 and 12). The extracted natural polysaccharide and the resulting microcapsules were characterized by different experimental techniques. Fourier transform infrared spectroscopy analysis of the CM showed the presence of uronic acid units and sugars. Scanning electron microscopy revealed that most particles were adhered together, causing the formation of compact, linked agglomerates, which resulted in different microstructures with irregular shapes. All oil–core microcapsules were characterized, and the results showed that the different shell materials could be used to microencapsulate sunflower oil. Among them, the microcapsule crosslinked with CM and Chi was the most suitable, with the highest encapsulation efficiency (95 %). This coacervation led to the narrowest size distribution of the capsules, with diameters ranging from 1 to 5 μm. Optical microscopy confirmed the deposition of coacervate droplets around oil drops and clearly showed that the formation of coacervated particles and their deposition onto oil droplets were successive events.

Keywords: *Opuntia ficus indica*; extraction; microcapsules; mucilage cactus.

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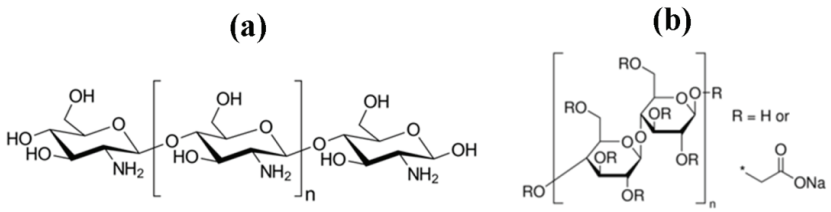
INTRODUCTION

Encapsulation is a technique allowing the inclusion of solid, liquid or gaseous substances within a support material.[1] It leads to the formation of particles. This process appeared in the early 1950s in the food industry. Griffin patented a process for the preparation of oils in the form of solids.[2] In 1956, Green and Schleicher[3] prepared pressure-sensitive capsules for the production of carbonless paper by complex coacervation between gelatin and gum arabic. Thanks to this work, they are now considered to be the pioneers of microencapsulation. Among these, microencapsulation using coacervation methods has become one of the most attractive alternatives in the cosmetics industry, as well as in the pharmaceutical, agro-chemical and food industries.[4–9] This process may be either simple or complex. In the simple coacervation process, a single polymer is involved. However the interaction of two or more oppositely charged polyelectrolytes in aqueous form, usually proteins and polysaccharides, leads to complex coacervate formation and phase separation. Microencapsulation by coacervation is establish in three steps made under continuous agitation. The first step concerns the formation of three immiscible chemical stages namely, the assembly liquid, the core medium, and the coating material. The second one, consists of a coating deposition stage, when the core material is dispersed in a polymer coating solution. The last one is the coating rigidification, when the immiscible material becomes more rigid and strong, which generally concerns warm, cross-connect, or desolvation methods. In all cases, the size of the microcapsules could be in the region of 1–5000 μm. Each capsule has two phases, namely the core and the coating or matrix material. The release mechanism of active compounds could be as following: *i*) dissolution-controlled released dissolution of the matrix or encapsulated dissolution system; *ii*) diffusion-controlled monolithic system nonporous/microporous matrix; *iii*) diffusion-controlled reservoir system nonporous/microporous matrix; *iv*) ion-exchange-controlled release. Therefore, the choice of the appropriate wall material is an extremely important factor for a successful coacervation process. Indeed, the microcapsule wall material must protect the encapsulated substance from deterioration, must have the required mechanical strength, must allow controlled release, and it must have thermal properties compatible with the encapsulated substance.[6,10]

Polysaccharides, which are the most abundant biomolecules,[7,11–15] have been used as microencapsulation matrices due to their good solubility in water and their low viscosity, even at high concentrations.[10,11] Several researchers reported a large number of publications on microcapsules based on polysaccharides.[6,10] Chitosan (Scheme 1a) is the polysaccharide most widely used as a membrane for microcapsules due to its biocompatibility and biodegradability properties as well as its antimicrobial activity. It is a commercially available polysaccharide produced from crustacean waste. The main field of application of mic-

rocapsules based on natural polymer is biomedical, which explains the numerous studies on the salting-out properties of the active ingredients according to the characteristics of the capsule or the external environment. The most commonly used production process is layer-by-layer deposition (LbL) on a core that can be removed later. Given the large number of publications on this subject, we have chosen to highlight only systems with a certain particularity. Another polysaccharide namely, CMCNa, is cellulose derivative with good solubility in water. The derivative is obtained by substitution reaction of hydroxyalcohol groups by hydrophobic carboxymethyl groups at position 2, 3, and 6 (Scheme 1b). CMCNa presents a linear polymer chain containing 1,4-linked glycans and displays polyelectrolyte behavior due to weak acidic groups in the molecular backbone. The substitution degree (DS) is one of the major factors affecting the physicochemical behavior of CMCNa, which consequently leads to its many diversified applications, such as thickener, emulsion stabilizer, fat replacer in meat, binder, and film-forming properties.[16–18] Moreover, concerning the safety of human health and the environment, CMCNa is biocompatible, biodegradable, and nontoxic.



Scheme 1. a) Chitosan; b) CMCNa.

The mucilage extracted from the cladodes of OFI is a hetero-polysaccharide with a molecular weight varying from 2.3×10^4 to 3×10^6 g mol^{-1}, being a complex mixture of polysaccharides, L-arabinose (24.6–42 %), D-galactose (21–40.1 %), D-xylose (22–22.2 %), L-rhamnose (7–13.1 %) and D-galacturonic acid (8––12.7 %).[19,20] Moreover, the neutral mucilage polysaccharide extracted from OFI using ethanol as the extracting solvent is considered non-toxic and safe for human and animal consumption.[21–23]

The main advantage of natural polymers is their biocompatibility and biodegradability making them more easily usable for pharmaceutical or medical applications. In addition, due to the growing demand of biobased materials for future sustainability, chitosan and carboxymethyl cellulose based material and their applications will bring good future perspective in fundamental and applied aspects of new product formulations. Both chitosan and carboxymethyl cellulose present several properties which are affected by a change in pH, such as swelling behavior, which allows increasing the matrix volume of polyelectrolyte and polyelectrolyte complex upon changing the pH from acidic to basic conditions. In this

context, the present study was conducted to evaluate the viability of the formation of microcapsules using different shell materials (CM extracted from OFI, carboxymethyl cellulose sodium salt (CMCNa), and chitosan (Chi)). Microencapsulation was performed using a segregative coacervation method.[20] The interactions and the stoichiometry between the biopolymers used in complex coacervation, as well as the ratio between wall materials, are important parameters in the encapsulation process. Another goal of this study was to evaluate the effect of cross-linking on the morphological characteristics, particle sizes, and encapsulation efficiency.

EXPERIMENTAL

Materials

Mucilage powders extracted from OFI were used as wall materials to prepare oil microcapsules. The cladodes were harvested from the oasis of El-Metkides-Gafsa, southwest of Tunisia. The epidermal cells of the OFI were removed and washed several times with distilled water to remove sand and hydrophilic impurities. The cleaned OFI was cut into chips (1×1×1 cm^3).[24]

Sunflower oil (palmitic acid: 5–7.6 %; stearic acid: 2.7–6.5 %; oleic acid:14–39.4 %; linoleic acid 48.3–74 %) was kindly donated by the SAS PIVERT (Compiègne, France). All other chemicals used (sodium hydroxide (NaOH, 98 %), hydrochloric acid (HCl, 37 %), carboxymethyl cellulose sodium salt (CMCNa), chitosan (degree of deacetylation: 75 %) and a cationic surfactant (tetradecyltrimethylammonium bromide, TTAB, 99 %, CMC = 4–5 mmol L^{-1})) were obtained from Sigma–Aldrich and used without further purification.

Mucilage extraction

The obtained chips were homogenized using 1 L of distilled water per 1 kg of material. Subsequently, the mixture was mechanically stirred at 80 °C for 20 min to inactivate enzymes and left to cool to ambient temperature. After cooling, the suspension was filtered to separate the liquid from the solids, and the obtained suspensions were centrifuged. The pH of the obtained filtrate was fixed at the desired pH value by the addition of a solution of hydrochloric acid or sodium hydroxide. Then, the mucilage was recuperated by precipitating the filtrate with ethanol. Then, in order to accelerate the precipitation, the obtained solution was centrifuged, and, finally, the mucilage was dried at room temperature for 3 days to obtain a powder, which was stored at 4 °C until further use. All the experiments were performed in triplicate.

Microcapsule formation

Oil–core microcapsules were prepared by simple and complex coacervation methods. The first step of the microencapsulation process consisted of forming an oil-in-water emulsion. This emulsion was prepared from a continuous aqueous phase and a dispersed oil phase. The continuous phase was obtained by dissolution of TTAB in 100 mL of water under magnetic stirring at 600 rpm. Both phases were then vigorously dispersed with an ultra turrax high speed homogenizer (Ika T 25 basic, France) at room temperature (≈20 °C) and 11000 rpm for 15 min to create an oil in water emulsion. During the formation of surfactant stabilized emulsion, the oil–water interfaces must be covered with an adequate number of particles to stabilize the droplets by providing repulsive interactions between drops and lowering the interfacial tension. The final surfactant concentration is therefore at their corresponding *CMC* (about four times the *CMC* value).

For simple coacervation, particles were obtained by the dropwise addition of an aqueous solution CM into the prepared emulsion. The pH of the premix was then adjusted to 4.0 by adding hydrochloric acid, 0.1 mol L^{-1}, in order to cross-link the capsules. Finally, the mixture was stirred mechanically at room temperature for over 2 h. The upper aqueous-rich phase was removed and the coacervated rich phase was recovered and stored for further analyses.

The complex coacervation was basically the same as the simple coacervation except that the CM and CMCNa solutions were added to the pre-emulsion vessel.

In order to crosslink the capsules prepared with cactus mucilage and chitosan (CM/Chi), a Chi solution was prepared by dissolving 5 mg of polymer in 100 mL of 2 % aqueous acetic acid solution at pH 4. CM/Chi coacervation was obtained by the dropwise addition of CM and Chi solutions (pH 3.6) in the required proportions. Then, the CM solution was added, and coacervation between the oppositely charged Chi and carboxylic groups of the CM started to form a thin coating layer around the core material. After 30 min, a 1 mol L^{-1} sodium hydroxide solution was added and the pH was adjusted to 8 to complete the coacervation reaction. All the experiments were performed in triplicate.

Characterization

Fourier transform infrared spectroscopy (FTIR) was used in this study to identify functional groups of the prepared CM powder. The spectra of the samples were recorded in the range of 4000–500 cm^{-1} in KBr pellets using a Shimadzu 8400S spectrometer (France) with a resolution of 4 cm^{-1}.

The particle size distribution of the powdered CM, the emulsions, and the suspensions of microcapsules was investigated by laser diffraction using a Mastersizer X Malvern (laser Cilas 1190, France). The sample was dispersed in aqueous medium under mechanical stirring for 2 min. The technique is based on measurement of the intensity of light scattered as a laser beam passes through a dispersed particulate sample. The test was repeated at least in triplicate and the difference between the various values obtained was within an experimental error of 5 %.

Scanning electron microscopy (SEM) was used to investigate the morphology of the different power microcapsules. The micrographs were recorded with a Zeiss-Ultra 55 SEM device, operating at 10 kV. The dried samples were fixed on SEM stubs using a double sided tape and coated with gold metal under high-vacuum evaporator.

The particle shape and surface morphology of the obtained particles were examined using optical microscopy (Axio Imager, Zeiss, Marly-le-Roi, France).

Carboxylic groups in the CM powder obtained at different supernatant pH values were quantified by conductometric titration. The CM sample (30 mg) was added to a beaker. and 200 mL of distilled water was added. The obtained solution was stirred for 10 min and ultrasonicated in order to obtain a well-dispersed suspension. Conductometric titration was performed using a 0.01 mol L^{-1} NaOH or HCl solutions. The number of functional groups is given by the following equation:

$$X = \frac{C(V_2 - V_1)}{w} \quad (1)$$

where X / μmol g^{-1} is the total acidic group content, C / μmol L^{-1} is the concentration of the sodium hydroxide solution, V_1 and V_2 in L are the amounts of NaOH or HCl, and w / g is the oven-dried weight of the sample. The test was repeated at least in triplicate and the difference between the various values obtained was within an experimental error of 5 %.

The encapsulation efficiency (*EE*) was evaluated according the procedure reported by Bae and Lee,[25] and Carneiro *et al.*[26] Fifteen milliliters of hexane were added to 1.5 g of pow-

der in a glass jar with a lid, which was shaken by hand for the extraction of free oil, during 2 min, at room temperature. The solvent mixture was filtered through a Whatman filter paper No. 1 and the powder collected on the filter was rinsed three times with 20 mL of hexane. Then, the solvent was left to evaporate until constant weight. The non-encapsulated oil (surface oil) was measured by weight difference between the initial clean flask and that contained the extracted oil residue. The total oil was assumed to be equal to the initial oil, since preliminary tests revealed that all the initial oil was retained, which was expected, since flax seed oil is not volatile. *EE* was calculated from the following equation:

$$EE, \% = 100\frac{\text{Total amount of oil} - \text{Surface oil}}{\text{Initial amount of oil}} \quad (2)$$

The test was repeated at least in triplicate and the difference between the various values obtained was within an experimental error of 5 %.

RESULTS AND DISCUSSION

Physicochemical characterization of mucilage powder

The FTIR spectrums of CM samples (Fig. 1) revealed typical absorption bands of polysaccharide between 800 and 1200 cm^{-1}, which is overpowered by ring vibrations overlapped with the characteristic vibrational modes of the pyranose ring.[27–29]

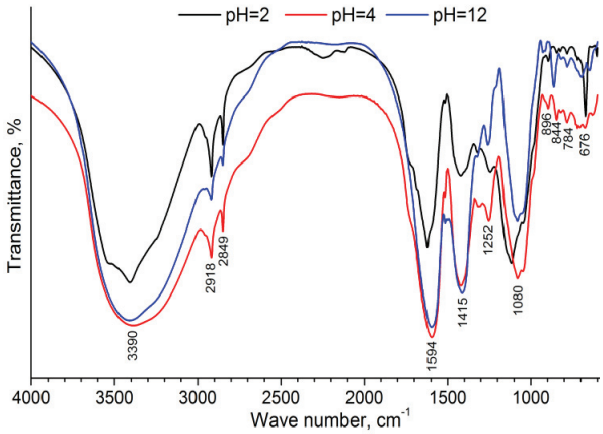


Fig. 1. FTIR spectra of different CM samples.

The absorption bands at 896 and 844 cm^{-1} indicate that the two configuration (α- and β-D-glucose) exist and that α-D-glucose was the major linkage.[27,30] The bands at 784 and 676 cm^{-1} are attributed to out-of-plane N–H and O–H vibrations, respectively.[31] The adsorption bands at 1252 and 1080 cm^{-1} result from vibration bonds C–O, C–C, O–H and C–O–C of the mucilage molecules.[32–34] Indeed, the wave numbers at 1594 and 1415 cm^{-1} are attributed to asymmetric and symmetric stretching vibration of deprotonated carboxylic acid groups (COO^-), probably due to the presence of uronic acid in CM samples.[31–36] In

addition, the band around 3390 cm^{-1} reflects the stretching vibration of the hydroxyl groups (–OH) and amine groups. While, the absorption bands around 2918 and 2849 cm^{-1} were assigned to asymmetric vibrations of C–H and methoxyl groups (CH_3O) in the CM samples.[32,33,37]

A wide distribution of particle size, as well as irregular morphology and different heterogeneous microstructures of the CM were observed by SEM (Fig. 2).

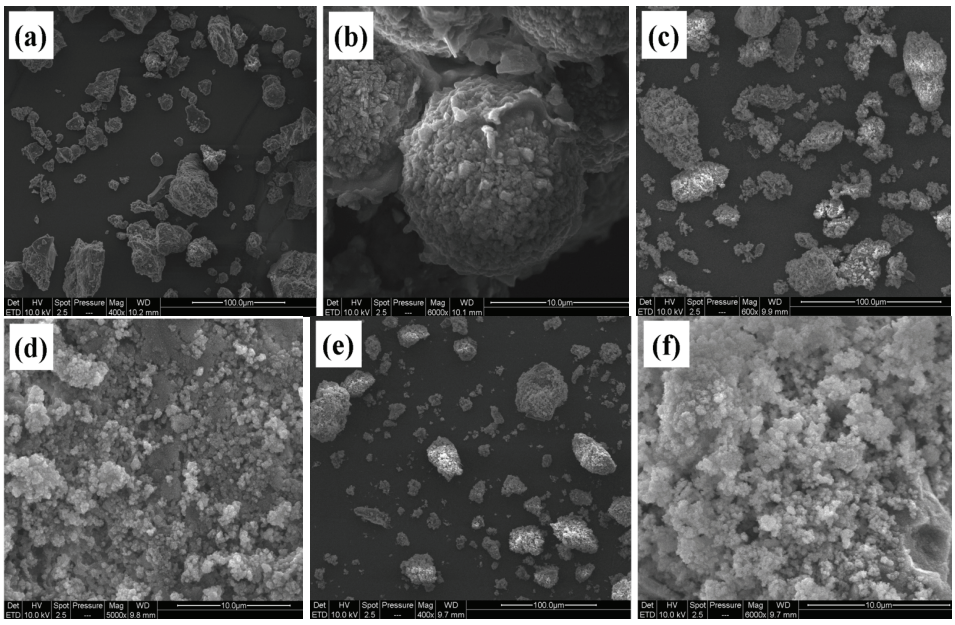


Fig. 2. Micrographics of CM powder ethanol extracted and precipitated at different pH values, 2 (a, b), 4 (c, d) and 12 (e, f).

The strong aggregation observed in the mucilaginous extracts obtained from OFI can lead to the formation of compact, bound agglomerates. Mucilaginous extracts obtained at pH 4 and 12 included various microstructures. The particles were interpenetrated at random in the entangled matrix of the polysaccharide chains. The agglomeration could be attributed to the effects of ionic crosslinking by calcium ions initially present in the sample. Indeed, Ca^{2+} and other metal ions naturally present in the CM[38] can enhance the ionic interactions between the dissociated carboxyl groups. The strong aggregation observed in the CM obtained from OFI agrees with that reported by Du Toit *et al.*[39] However, an obvious difference and incomplete crosslinking was observed in the microstructure of the CM obtained at pH 2. This CM had a spongy structure of spherical aggregates of irregular shape at different sizes, formed by the adhesion and superposition of polymeric chains. These observations suggest that chelation has a significant

effect on morphology and that metal ions can contribute to the formation of compact, bound agglomerates with sufficient dissociated carboxyl groups.[40]

The average particle size, as determined by laser diffraction analysis, was established and the results are shown in Fig. 3. The dried CM powder had almost the same particle size distribution and consisted of fine particles within the range 46 to 595 μm. The presence of very small particles facilitated a greater number of particle-to-particle interactions and, subsequently, the formation of agglomerates. The polysaccharides obtained at basic pH contained the highest number of carboxyl groups, and the number of carboxylic groups depended on the pH. Thus, the concentrations of carboxylic groups were 0.733, 0.870 and 3.033 μmol g^{-1} for the CM obtained at pH 2, 4 and 12, respectively.

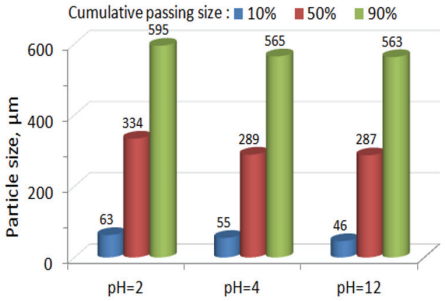


Fig. 3. Particle size distribution of the CM samples.

Physicochemical characterization of the emulsion and microcapsules

Particle size is an important parameter in microencapsulation because it influences the texture and sensory aspects of particles and it defines their applications. The stable, primary emulsions of oil were used for the preparation of the microcapsules by different coacervation methods. After addition of surfactants, stable oil-in-water emulsions were successfully prepared using CMC concentrations. The average particle sizes of the emulsions ranged from 0.45 to 4 μm (Fig. S-1 of the Supplementary material to this paper). The size distribution curves (Fig. S-1) showed unimodal and homogenous distributions (the diameters were distributed in a single peak). An image of these emulsion drops is shown in Fig. S-1 showing bleaching of the aqueous phase caused by displacement of the particles from the drops to the continuous phase. These emulsion drops remained stable for at least 6 months with no measurable change in the size distribution of the drop. This due to the ability of TTAB surfactant to minimize the energy required for emulsion formation by reducing the liquid–liquid interfacial tension and the formation of electrostatic or steric barriers around the droplet surfaces.

The average diameters of the microcapsules obtained by simple coacervation ranged from 50 to 400 μm (Fig. 4), which confirms that there was flocculation on the surface, with a hard and difficult to break shell. This could be associated with

the process conditions, including emulsion preparation, the cooling rate, and the characteristics of the wall and core materials.

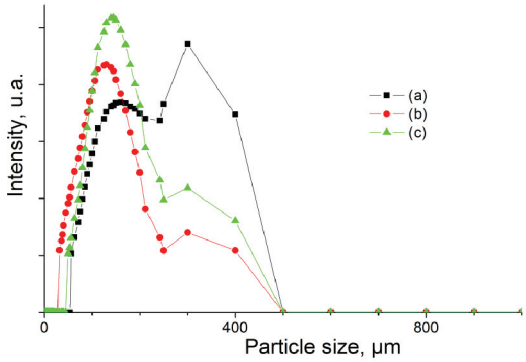


Fig. 4. Particle size distribution of microcapsule suspensions *via* simple coacervation at pH 4 crosslinked by CM precipitated at pH: 12 (a), 4 (b) and 2 (c).

Likewise, increasing the core-to-wall ratio increases the size of the microcapsules and their distribution. Increasing the proportion of encapsulating polymer or reducing the velocity of stirring resulted in increasing microcapsule size.

However, despite the small difference observed in the particle size distribution of the emulsions and the corresponding suspensions of microcapsules prepared using CM/CMCNa and CM/Chi by complex coacervation (Figs. 5 and 6), the microcapsules were successfully prepared, and were stable. This is probably related to the increase in the number of crosslinks, leading to strengthening of the walls of the microparticles.[41] Thus, the addition of the anionic polymer CMCNa with the CM solution to the prepared emulsion results in electrostatic repulsion between the biopolymers, leading to phase separation and coacervate formation. Subsequently, these coacervates form deposits around the sunflower oil droplets.

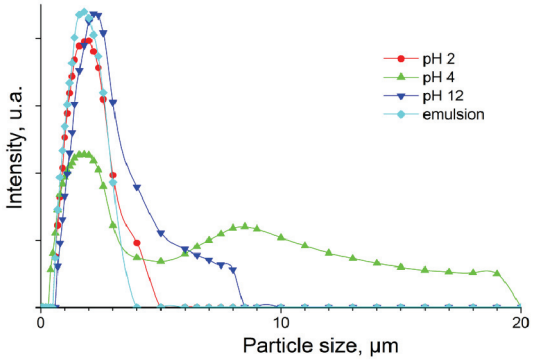


Fig. 5. Particle size distribution of emulsions and microcapsule suspensions *via* complex coacervation at pH 4 crosslinked with CMCNa and CM precipitated at different pH values.

A slight variation was observed between the emulsions and the corresponding suspensions of microcapsules, especially with the use of Chi. This lead to a much narrower size distribution, with diameters that ranged from 1 to 5 μm. The

ability of Chi and CM to form a very thin coating layer can be explained by the electrostatic attractions between oppositely charged biopolymers. These interactions induce the formation of complexes, which can be insoluble, and they are closely related to the pH of the two solutions, as well as the weight ratio of the CM/Chi mixtures. Chi developed a positively charged surface in the acidic aqueous medium due to the presence of amino groups, and CM displayed a net negative charge.[42]

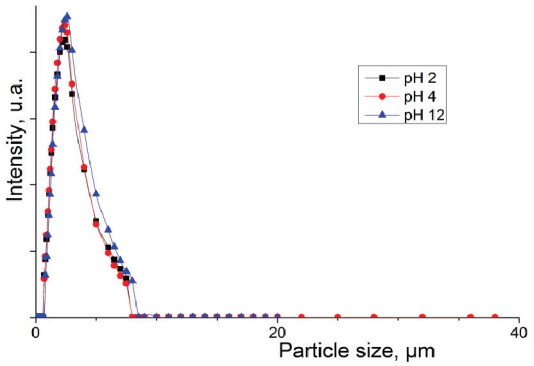


Fig. 6. Particle size distribution of microcapsule suspensions *via* complex coacervation precipitated at pH 8 and crosslinked with Chi and CM obtained at different pH values.

The electrostatic interactions in the aqueous system between the protonated Chi amino groups and the negatively charged COO– groups of CM were due to the formation of a stable polyelectrolyte complex at pH 8. Therefore, a barrier was built around the sunflower oil by the formation of additional functional groups, primarily azomethin linkage groups (>C=N<) and, secondly, acetal linkages (–C–O–C–O–C–).[43] The particle size of the obtained microcapsules was confirmed by optical observation. Thus, microcapsules were prepared by simple and complex coacervation *via* mucilaginous cactus extracts. It is clear that the active ingredient was well surrounded and protected by the coacervate deposits (Fig. S-2 of the Supplementary material).

Optical observation analysis revealed that the microparticles obtained by complex coacervation were round, with various sizes and a polynuclear distribution of the core material (Figs. S-3 and S-4 of the Supplementary material). These results were related to the different cactus extract compositions and their influences on the diffusivity and film formation properties of these extracts.

The effect of wall material on encapsulation efficiency (EE) of sunflower oil is recapitulated in Fig. 7. It could be observed that the encapsulation efficiencies of the microcapsules obtained by complex coacervation and crosslinked with Chi and CM are different than those of the system CM/CMCNa. For the former system. CM/Chi, the EE values ranged from 87 to 95 %. Whereas, the encapsulation efficiencies for the latter system, CM/CMCNa, varied from 65 to 72 %. It could be concluded that it is possible to use the different cactus extracts as carrier mat-

erials to protect sensitive compounds. The results also showed that the suggest methodology (complex coacervation *via* CM/Chi) and the chosen encapsulation polymers were adequate and very effective for the proposed goal.

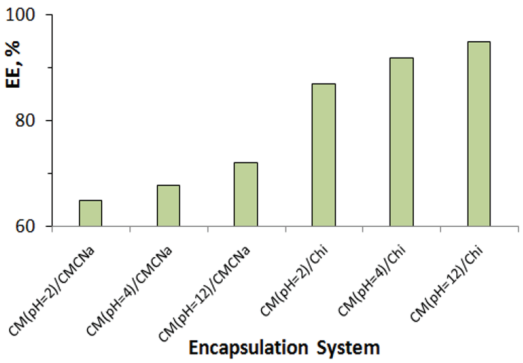


Fig. 7. Effect of wall material on encapsulation efficiency (*EE*) of sunflower oil.

CONCLUSIONS

This work consisted of developing bio-based microcapsules using CM and CMCNa as well as chitosan. which are the polysaccharides most commonly used in virtually all the reported microencapsulation methods. It can be highlighted from the results that CM is a promising agent for the microencapsulation of bioactive molecules. The microcapsules crosslinked with Chi and different cactus extracts had sufficient stabilities needed to maintain their structures, with good entrapment efficiency (87 to 95%) compared to those crosslinked with CMCNa or only with CM. The morphologies and particle sizes of the resultant microcapsules were influenced by the polymer type. Optical observation confirmed that core–shell morphology existed in all types of capsules, with spherical morphologies and smooth surfaces. The integrated data indicated that the sunflower oil was successfully encapsulated by complex coacervation using the cactus extract as the carrier material. Moreover, CM could be used as a potentially effective shell material, especially with a cationic polymer, for protection of bioactive compounds from oxidative deterioration and for drug delivery.

SUPPLEMENTARY MATERIAL

Additional data are available electronically at the pages of journal website: https://www.shd-pub.org.rs/index.php/JSCS/index, or from the corresponding author on request.

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ИЗВОД

ПРИМЕНА СЛУЗИ ИЗОЛОВАНЕ ИЗ КАКТУСА *Opuntia ficus indica* као ОПНЕ ЗА МИКРОИНКАПСУЛАЦИЈУ

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У раду је испитано стварање микрокапсула користећи природне биополимере, као што су слуз кактуса (CM), натријумова со карбоксиметил целулозе (CMCNa) и хитозан (Chi), за транспорт и доставу уља сунцокрета. CM узорци су екстраховани из *Opuntia ficus indica* (OFI) таложењем супернатанта на различитим pH (2, 4 и 12). Екстраховани природни полисахарид и формиране микрокапсуле су окарактерисане различитим методама. Инфрацрвена спектроскопија са Фуријеовом трансформацијом (FTIR) CM је указала на присуство јединица уронске киселине и шећера. Скенирајућа електронска микроскопија (SEM) је показала да је већина честица атхерирала стварајући компактне агломерате, дајући различите микроструктуре неправилних облика. Резултати су показали да се могу користити опне од различитог материјала за инкапсулацију уља сунцокрета. Највећу ефикасност су исказале микрокапсуле са умреженим CM и Chi (95 %). Ови коацервати су имали најмањи опсег величине капсула, пречника од 1 до 5 μm. Оптичком микроскопијом је потврђено окруживање капи уља капима коацервата.

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