

Protective action of lemongrass essential oil on mucilage from chia (*Salvia hispanica*) seeds

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A B S T R A C T

Mucilage extracted from chia seeds (*Salvia hispanica*) is reckoning an increasing interest for its mechanical and nutritional properties. Freeze-dried mucilage dispersed in solvent undergoes degradation, through autoxidation phenomena, that results in the loss of the initial rheological characteristics. In order to limit the extent of depolymerization, an essential oil (lemongrass essential oil, LEO) is here emulsified with chia mucilage (CM) suspensions, and the rheological behavior of samples containing LEO or not compared. Flow curves of CM suspensions and of CM emulsions dispersing LEO (O/W-CM) demonstrated that both the systems can be classified as plastic fluids, and that during storage a general decrease of the viscosity of CM suspensions is detected, while the viscosity of the O/W-CM systems remaining constant. The differences between the two systems were also confirmed by oscillatory tests and through measurements of intrinsic viscosity. By means of an accelerated oxidative test, it was unveiled that the short-term stability of CM suspensions is attributable to the reaction of oxidation, and that the presence of LEO, due to its anti-oxidative properties, allowed for preventing degradation phenomena on the chia mucilage.

We conclude that the colloidal stability of chia-based mixtures can definitely be improved following to LEO treatment - hence forming an emulsion - thereby preventing short-term polymer degradation.

1. Introduction

Hydrophilic polymers dispersed in water form suspensions with the properties of colloids, hence assembling hydrocolloids. This kind of materials finds application in several fields ranging from biomedical to food technologies (Goff & Guo, 2020; Vecchies et al., 2018; Williams & Phillips, 2009). In food industry, hydrocolloids are used as gelling agents or thickeners since they are able to retain discrete amount of water (Saha & Bhattacharya, 2010) and to modify the food structure. The study of the rheological characteristics of hydrocolloids or hydrogels is central for their use in food processing since the different flow behavior and viscosity can affect the fluid handling in plants. The most interesting hydrocolloids in food industry encompass natural polymers that offer a versatile degree of flexibility for many applications (Cuomo, Cofelice, & Lopez, 2019; Perugini et al., 2018). Among them, sodium alginate, agar, carrageenan, locust bean gum, guar gum, gum arabic, xanthan gum,

gellan gum are used in food preparations as additives (Smith & Hong-Shum, 2003).

Hydrogels are known as water-swollen cross-linked polymers characterized by covalent bonds or physical interactions endowed with peculiar mechanical properties (Cok et al., 2018; Sacco, Cok, Asaro, Paoletti, & Donati, 2018). Many polymer-based matrices reported in the literature are obtained by polymer dispersions, where the use of specific gelling agents represents a mandatory step to form covalent or ionic bonds (Cuomo et al., 2019). Recently, Samateh and colleagues (Samateh et al., 2018) demonstrated that hydrogel-like structures can be assembled in absence of external cross-linkers by some seeds like those of chia (*Salvia hispanica*) and basil (*Ocimum basilicum*) when soaked in water, hence forming a hydrogel-like matrix through an extensive network of nanoscale fibers protracted from the seeds surface into the aqueous bulk. The set of secreted fibers from the seeds is indicated in various studies as mucilage.

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Salvia hispanica is an annual herbaceous crop species of the Lamiales family, native in Mexico and Guatemala (Whistler, 1982). Presently, the nutritional assets of chia seed have been valued and its health benefits documented (Ding et al., 2018; Muñoz, Aguilera, Rodríguez-Turiénzo, Cobos, & Diaz, 2012; Scheer, 2011; Valdivia-López & Tecante, 2015). Due to the benefits coming from the presence of ω -3 and ω -6 fatty acids, vitamins, antioxidants and minerals chia seeds constitute an ingredient in food industry applications (Orona-Tamayo, Valverde, & Paredes-López, 2017; Soukoulis, Gaiani, & Hoffmann, 2018; Taga, Miller, & Pratt, 1984; Weber, 2000).

Chia seeds, besides being a source of oil and protein, are certainly a source of polysaccharides (Bushway, Belyea, & Bushway, 1981), and for this reason they represent suitable candidates for the fabrication of emulsion systems in food industry. Muñoz and colleagues (Muñoz, Cobos, Diaz, & Aguilera, 2012) studied the mucilage release from the seed coat during hydration, finding that it can be easily extracted and hydrated to achieve water retention of 27-fold its weight, thus underlying the great potential as a functional ingredient to be used as a thickener in foods. Since then, an increasing interest toward the mucilage has been noticed in literature (Capitani et al., 2015; Capitani, Ixtaina, Nolasco, & Tomás, 2013; Segura-Campos, Ciau-Solís, Rosado-Rubio, Chel-Guerrero, & Betancur-Ancona, 2014; Timilsena, Wang, Adhikari, & Adhikari, 2016).

Salgado-Cruz et al. (Salgado-Cruz et al., 2013) characterized the chia seeds and the mucilage produced from the seeds when soaked in water. The authors, through microscopic imaging, observed the seed hydration and the release of the mucilage. The composition of the chia mucilage has been reported by Coorey et al. (Coorey, Tjoe, & Jayasena, 2014) that determined on the freeze-dried gel foam the contents of moisture, ash, protein, fiber, oil, and fatty acid profile. In particular, they found a content of 58% of crude fibers (insoluble fraction) and 35% of carbohydrates. Goh et al. (Goh et al., 2016) investigated on the effect of the temperature on the chia gel that was sensitive to changes of this parameter as well as to changes of pH, ionic strength and to the type of cation. They also determined the mucilage composition finding a slightly higher crude fiber content (65%) (Goh et al., 2016), a difference that was probably due to the differences in the extraction method.

For its rheological and nutritional characteristics, lately, chia mucilage has been proposed as fat replacer in different food preparations like bread and cakes (Felisberto et al., 2015; Fernandes and de las Mercedes Salas-Mellado, 2017) and in dressing sauces (Fernandes & de las Mercedes Salas-Mellado, 2018). Moreover, some attempts have been made for the production of edible films based on chia mucilage together with chia seed protein and clove oil (Capitani et al., 2016) and together with glycerol (Dick et al., 2015).

A drawback in the use of some hydrocolloids is the occurrence of the autooxidation phenomena consequently to the formation of free radicals (Gryn'ova, Hodgson, & Coote, 2011). This will reduce the molecular weight of the polymer, thus creating modifications of the mechanical and organoleptic properties. Therefore, there is a great demand of strategies able to slow down the auto-oxidative processes.

Among antioxidants, essential oils represent a class of natural compounds (Amorati, Foti, & Valgimigli, 2013) widely accepted by consumers and in some recent studies lemongrass essential oils (LEO) have been emulsified in polymer matrices in order to provide a primary or a secondary packaging to food commodities (Cofelice, Cuomo, & Lopez, 2018; Cofelice, Lopez, & Cuomo, 2019; Raybaudi-Massilia, Mosqueda-Melgar, Soliva-Fortuny, & Martín-Belloso, 2016; Salvia-Trujillo, Rojas-Graü, Soliva-Fortuny, & Martín-Belloso, 2013).

The aim of this study was to investigate on the effects of the LEO addition to chia mucilage suspensions. Through rheological measurements during samples storage, the differences between chia mucilage suspensions and the chia mucilage emulsified with LEO are described. Moreover, the protective role played by LEO in chia stability is also disclosed by means of oxidation reaction tests.

2. Materials and methods

2.1. Materials

Chia seeds (Salba grain organic) were obtained from I.P.A. s.r.l. Industria Prodotti Agroalimentari (Viterbo, Italy), lemongrass (*Cymbopogon nardus*) essential oil (100%) was from Erbae (Lama di San Giustino -PG - Italy), sunflower oil was purchased in a local supermarket. 2,2'-Azobis(2-amidinopropane) dihydrochloride (AAPH) was from Wako Chemicals and sodium fluorescein from Fluka.

2.2. Mucilage extraction from seeds

Chia seeds were weighed and placed in a vessel with ultrapure water in a 1:20 (w/v) ratio for 12 h to extract the mucilage (CM). The CM was recovered through vacuum filtration with a Buchner funnel, frozen at -40°C and then freeze-dried under vacuum at 15 Pa in a freeze-drier Genesis 25 ES (VirTis, NY, USA) for 48 h (maximum shelves' temperature $+20^{\circ}\text{C}$). After freeze-drying, CM, with the aspect of a dried foam, was stored at room temperature.

2.3. Proximate analysis

Freeze-dried mucilage was analysed using ICC standard procedures (ICC, 1995) for moisture (Method 110/1), protein (N x 6.25) (Method 105/2), and ash (Method 104/1). Total fiber amount was determined using AACC Method 32.07 (AACC, 2000).

2.4. Mucilage suspensions preparation

Aqueous CM suspensions were prepared at different concentrations (from 0.025 to 1.8% w/w). The freeze-dried CM was dispersed in warm ultrapure water (50°C) under stirring until complete disappearance of macroscopic aggregates.

2.5. Preparation of mucilage emulsions (O/W-CM)

The oil in water emulsions were prepared at 25°C by dispersing LEO (or sunflower oil) in CM suspensions. The oil was emulsified (4 min at 24000 rpm through a homogenizer Ika T25 Digital Ultraturax, Staufen, Germany) in a small amount of CM suspension that was further mixed (through a magnetic stirrer) with an aliquot of non-homogenized CM suspension (CM had the same concentration as in the continuous phase of the dispersion).

2.6. Intrinsic viscosity measurements

The intrinsic viscosity $[\eta]$ of CM was measured at 25°C by means of an AVS®370 viscosity measuring system equipped with a CT 72/P thermostat (SI Analytics), using a Schott capillary viscometer. Deionized water was used as solvent (Timilsena et al., 2016). Freeze-dried CM was solubilized in deionized water at 60°C for 24 h. Next, CM suspension was incubated in the presence or not of LEO (CM:LEO 2:1 w/w). Samples were then centrifuged (1000 x g, 30 min) and resulting supernatants analysed at time 0 and after 7 days of incubation at room temperature and gentle stirring. Intrinsic viscosity was calculated by analyzing the polymer concentration dependence of the inherent viscosity, i.e. the reduced logarithm of the relative viscosity, $\ln(\eta_{rel})/c$ using the Kraemer equation (Eq. (1)):

$$\frac{\ln \eta_{rel}}{c} = [\eta] - k[\eta]^2 c \quad (1)$$

where k is the Kraemer constant, η_{rel} the relative viscosity and c the polymer concentration.

2.7. Rheology

Rheological measurements were carried out using a rotational rheometer (Haake MARS III-Thermo Scientific, Karlsruhe, Germany) equipped with a 60 mm parallel plate geometry probe (PP60). The instrument was equipped with a Peltier element combined with a Phoenix II digital system (Thermo Scientific, Karlsruhe, Germany) for the temperature control. Samples (2.9 mL) were left equilibrating for 5 min before measurements. Steady-shear flow tests were performed in controlled shear rate (CR) in the range of 0.01–200 s⁻¹ (Capitani et al., 2013). Oscillation strain sweep measurement were carried out for determining the linear viscoelastic range (LVE) at a fixed frequency, ν , of 1 Hz (McClements, 2011).

2.8. Oxidation test

CM oxidation was monitored as a function of time using a fluorimetric method. The reaction of oxidation was triggered by the thermal activation of AAPH and the reaction evolution was followed through the fluorescein emission (Ou et al., 2002). Briefly, chia mucilage suspension or emulsion were prepared at CM concentration of 0.2% and 0.1% of LEO. Before being incubated with the radical initiator and the fluorescent probe, the samples were diluted as to prevent the turbidity from interfering with the fluorescence measurement. AAPH was used to a final concentration of 50 μ M and fluorescein at 5 μ M. Systems loaded with fluorescein and radical initiator were placed in the spectrofluorometer (Varian Eclipse, Palo Alto, CA, USA), incubated at 45 °C and the emission spectra of fluorescein excited at 482 nm and emitting at 513 nm were recorded every 5 min.

2.9. Optical microscopy

The microscopic aspect of the o/w emulsions was evaluated through optical microscope (Optech B5T microscope) interfaced to a camera (Bresser Camera, MikrOkular Full HD, Explore Scientific GmbH, Rhede, Germany).

3. Results and discussion

3.1. CM extraction and composition

Chia seeds contain high amounts of polysaccharides, proteins, lipids and fibers (Felisberto et al., 2015). When placed in water, the seeds exude a gel-like mucilage predominantly composed of water soluble polysaccharides forming supramolecular complexes tightly interacting with the seed surface (Lin, Daniel, & Whistler, 1994). Here, exuded mucilage was separated from seeds through vacuum filtration, collected, freeze-dried and stored in dried conditions at room temperature. The yield extraction of the mucilage was about 5% (w/w) of the dried seed mass (calculated as the amount of freeze-dried mucilage to the amount of seeds). The proximate analysis of the freeze-dried mucilage is reported in Table 1.

3.2. Rheology of CM water dispersions

In order to study the physical properties, CM was dissolved in water

Table 1
Proximate composition (%) of freeze-dried chia mucilage.

Components	Composition % (\pm St.Dev)
Moisture	11.3 \pm 0.1
Total fiber	75.3 \pm 0.2
Proteins	9.7 \pm 0.1
Ash	6.8 \pm 0.2
Fat	n.d

at different concentrations ranging from 0.025% to 1.8% w/w. Steady-shear flow tests of the CM suspensions were performed and sample-case flow curves are reported in Fig. 1(a). As can be seen, for all the CM concentrations the shear stress (τ) increased with the increase of the shear rate ($\dot{\gamma}$) in a non-linear way (shear-thinning behavior), indicating a deviation from the Newtonian fluids behavior. The best fitting equation for all the flow curves was the Herschel-Bulkley model (Mezger, 2006) (Eq. (2)):

$$\tau = \tau_0 + k\dot{\gamma}^n \quad (2)$$

where τ is the shear stress, τ_0 is the yield stress, k is the consistency index, $\dot{\gamma}$ is the shear rate and n is the index of rheological behavior. According to eq. (2), the CM suspensions behave like plastic fluids and present a yield stress. The values of k and n as a function of CM concentration are reported in Fig. 1(b) and listed in Table S1 together with the correlation coefficients of the fittings to eq. (2). As inferred, values of k were small at low mucilage concentration (lower than 0.8%) and started to rapidly increase from 1 to 7 by increasing the mucilage concentration from 1 to 1.4%. Values of n decreased considerably reaching a value of 0.48 at mucilage concentration of 1.4%. Since values of n close to 1 are indicative of a Newtonian behavior and as they approach to 0 they move further and further away from the Newtonian behavior, there was an increasingly plastic behavior with the increase of the mucilage content.

Consistency index values were also measured for some CM concentrations at different temperatures (Fig. 1(c)). The decrease in the k values on temperature uprise indicated that the suspensions viscosity decreased with this variable, in line with the classic behavior of the polymers in aqueous solution (Mezger, 2006).

During the CM suspension storage, the formation of macroaggregates was noticed in the bulk phase having, at naked eye, the aspect of wires. It seemed like the water solvent expelled part of the CM. Physical changes of the CM suspensions were also confirmed by the acquisition of flow curves within a timeframe of 10 days. As can be seen from Fig. 2(a), (c) and (e), that show the flow curves of aqueous suspensions of CM at concentrations of 0.2, 0.4 and 0.6% respectively, after few days of storage the values of apparent viscosity started decreasing. This occurrence may be associated with the CM oxidation (*vide infra*), considering that the main cause of the degradation of biopolymers is the oxidative damage (Wellington, 1983).

3.3. O/W-CM oil in water emulsions

For the above reasons the effects of the addition of LEO, an oil with well-recognized antioxidant and antimicrobial properties (Raut & Karuppaiyl, 2014), was tested to prevent CM spoilage. LEO at the concentration of 0.1% w/w was mixed with the CM suspension, thus forming oil in water emulsions (O/W-CM) without the addition of any supplemental surface active agent and the flow curves were collected on O/W systems within the same timeframe as for the CM suspensions and a plastic flow behavior was detected also on O/W-CM emulsions. As displayed in Fig. 2(b), (d) and (f), the viscosity values of the emulsions did not change during storage thus indicating that essential oil had positive effects on the stability of the chia suspensions and showing the surfactant properties of CM, probably attributable to the protein fraction extracted from the chia seeds (see Table 1). Oil-in-water emulsions were also prepared using sunflower oil. The flow curves collected within time are illustrated in Fig. S1 and demonstrated that the use of sunflower oil did not stabilize the CM, unlike LEO. For this reason, emulsions with sunflower oil were excluded from further investigation. The aspect of the LEO based O/W-CM emulsion was investigated through optical microscopy and the micrograph acquired is reported in Fig. S2.

The viscoelastic characteristics of CM and O/W-CM systems were determined in oscillatory conditions. The amplitude sweep test presented in all the CM suspensions the storage modulus G' always higher

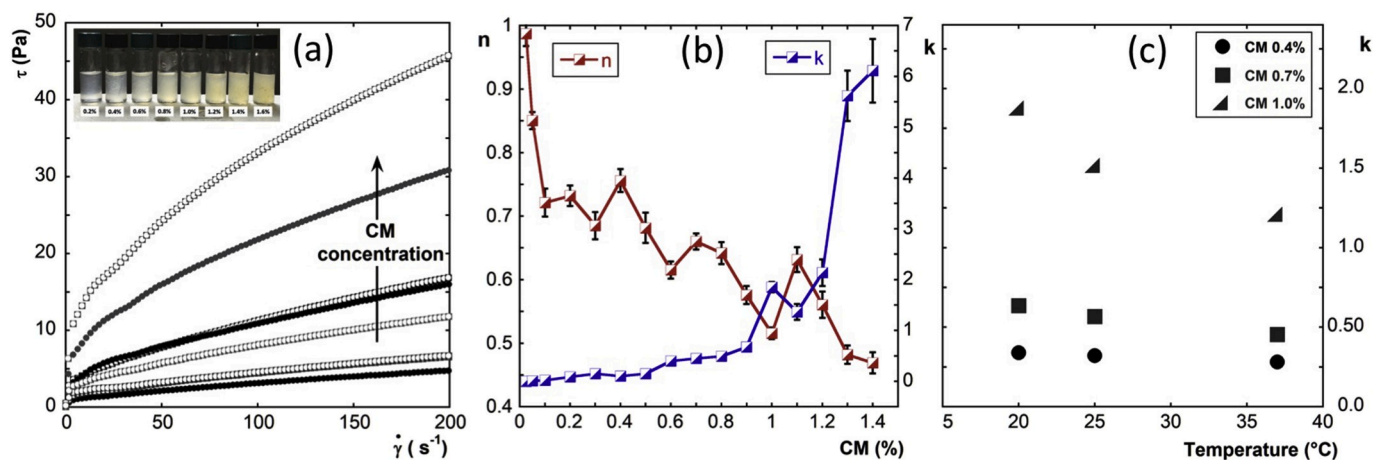


Fig. 1. (a) Shear stress τ as a function of the shear rate ($\dot{\gamma}$), inset of the figure shows the aspect of chia mucilage (CM) suspensions with the increase of CM concentration; (b) consistency index (k) and rheological behavior index (n) of CM as a function of the concentration; (c) consistency index of CM at concentrations of 0.4, 0.7 and 1.0% and at different temperatures.

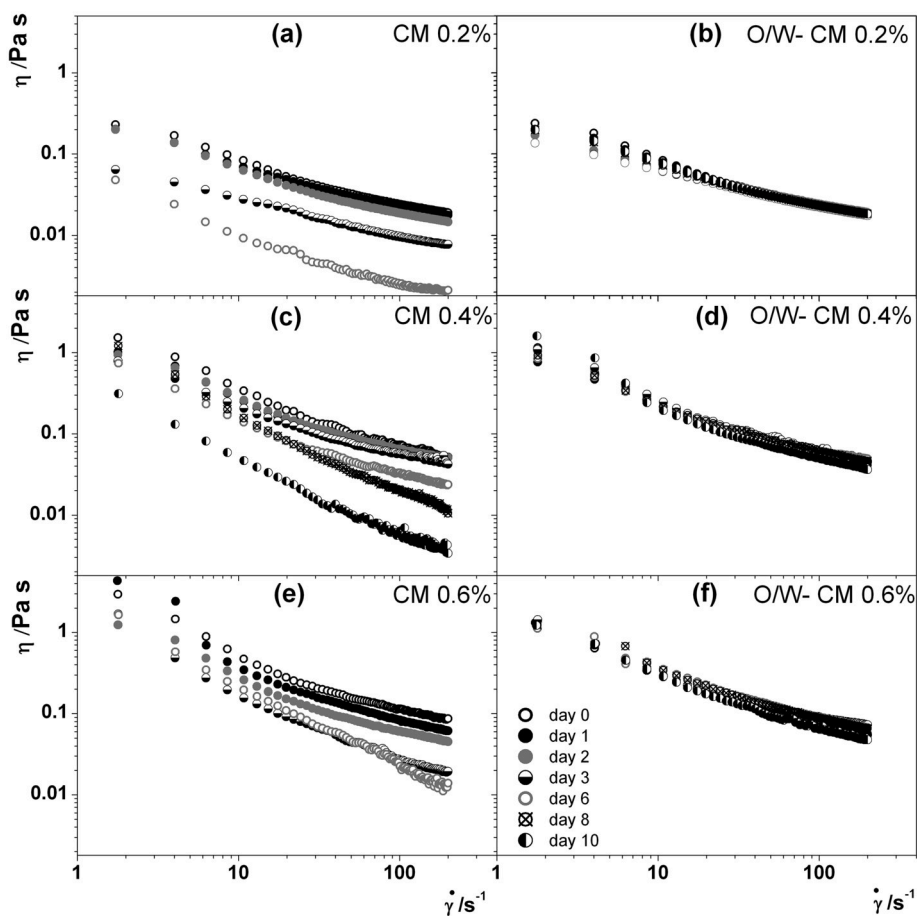


Fig. 2. Apparent viscosity curves of CM suspensions and CM based oil in water emulsions collected within 10 days. (a), (c), (e) viscosity curves of CM suspensions at concentration of 0.2%, 0.4% and 0.6%, respectively. (b), (d), (f) o/w emulsions with LEO concentration of 0.1% and CM concentration of 0.2%, 0.4% and 0.6%, respectively.

than the loss modulus G'' , thus indicating that the suspensions had a gel-like behavior (Fig. S4). The value of the elastic modulus, moreover, increased with the concentration of mucilage (Fig. S3). This result indicated that the strength of the polymeric interconnections increased together with the resistance of the material that became more solid-like according to the increase of CM concentration.

Fig. 3 (a) and (c) illustrate the G' modulus as function of the deformation within 10 days for CM at 0.4% and 0.6%, respectively. It is noticeable that the stability of the suspension decreased with time since the length of the plateau region of G' was shortened some days after the preparation. On the contrary, the response of G' remained constant during storage for the O/W-CM preparations as shown in Fig. 3 (b) and

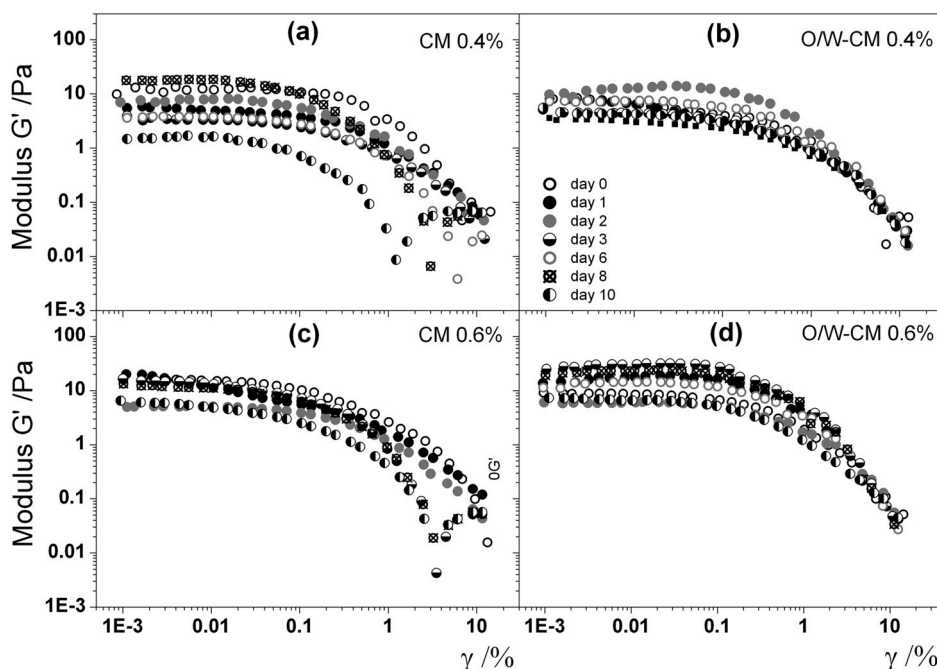


Fig. 3. Elastic modulus (G') as a function of the applied deformation (γ). (a), (c), mechanical spectra of CM suspensions at concentration of 0.4% and 0.6%, respectively. (b), (d) mechanical spectra of o/w emulsions with LEO concentration of 0.1% and CM concentration of 0.4% and 0.6%, respectively.

(d).

To further study the protective role played by the emulsified LEO, we next undertook intrinsic viscosity measurements in dilute conditions. Fig. 4(a) shows the inherent viscosity trend of CM suspensions and Fig. 4(b) that of O/W-CM systems determined by fitting the data to infinite dilution using Kraemer equation (eq. (1)). In the two graphs, the values of inherent viscosity were determined on the systems freshly prepared and after seven days from the preparation. The intrinsic viscosity values extrapolated at c_{-0} confirmed that in absence of essential oil CM undergoes degradation and, since part of the polymer is segregated from water, the value of intrinsic viscosity dwindled. On the contrary, the occurrence of this phenomenon is dramatically reduced in the presence of essential oil. In fact, by comparing (Fig. 4(c)) the intrinsic viscosity reduction values calculated for both the systems, we noticed a $[\eta]$ drop of approximately 80% in the case of pure CM suspension after 7 days of incubation whereas 30% for the O/W-CM system.

To in-depth understand the specific role of LEO in the storage of the CM, its antioxidant capacity was tested through the oxidation reaction triggered by the free-radical azo-initiator AAPH that produced peroxy radicals by thermal decomposition. The peroxy radicals chain reactions involved the CM as substrate and the radical species provoked the reduction of the fluorescein molecules resulting in the loss of the fluorescence emission of fluorescein as can be seen in Fig. 5 for samples containing CM only and water. In emulsions, the presence of LEO plays the antioxidant role shielding fluorescein from the reduction reaction thus slowing down the emission decay of fluorescein. Relative emission decay of fluorescein is shown in Fig. 5, where the persistence of the fluorescence emission as function of time is detectable in presence of LEO in O/W-CM emulsion, while a faster emission decay was detected in presence of CM suspensions and in water. This outcome confirmed that the CM is susceptible to the reaction of oxidation and that the essential oil can protect the mucilage from the peroxy radicals attack.

On the other hand, very recently, Hernández-Morales et al. (Hernández-Morales et al., 2019) have taken advantage of the susceptibility to the oxidation of the CM, whose principal polysaccharide is a tetrasaccharide composed of (1 \rightarrow 4)- β -D-xylopyranosyl-(1 \rightarrow 4)- α -D-glucopyranosyl-(1 \rightarrow 4)- β -D-xylopyranosyl units (Lin et al., 1994), for the green synthesis of Ag nanoparticles starting from silver nitrate. In

the proposed mechanism the synergic occurrence of the polysaccharide oxidation and the contemporary reduction of Ag^{+1} to Ag^0 were highlighted. Moreover, considering that the main part of the CM is made of insoluble fiber and soluble carbohydrates and that during storage in water the segregation of part of the suspended mucilage in macroaggregates is detected, it can be speculated that as soon as CM is dispersed in water, the insoluble fiber is held in suspension by the soluble polysaccharides. In this conditions when the latter undergo oxidation/degradation the insoluble fraction of the fiber undergoes precipitation determining the formation of macroaggregates.

4. Conclusions

This study has effectively shown that the presence of lemongrass essential oil supplied to mucilage extracted from chia seeds suspensions *via* the formation of an oil in water emulsion prevents oxidation phenomena on the chia mucilage solutions. The results were obtained through a rheological characterization supported by intrinsic viscosity measurements. The degradation of the macromolecule dissolved in water, through autoxidation phenomena was connected with the loss of the initial rheological characteristics. By comparing flow curves of chia mucilage suspensions and of mucilage based emulsions containing essential oil we can conclude that both the systems shall be classified as plastic fluids and that during storage a general decrease of the viscosity of chia mucilage suspensions is detected while the viscosity of the emulsion systems remained unchanged. These differences were also confirmed by oscillatory tests and through measurements of intrinsic viscosity. The chia based emulsion enriched with lemongrass essential oil here studied exhibited excellent stability against coalescence for long-term period.

To the best of our knowledge, this is the first time that chia mucilage has been used in the form of an emulsion. The majority of the studies so far available are focused on the characterization of the chia mucilage. The more recent ones are based on the application of chia mucilage as fat replacer in the preparation of bakery products (Felisberto et al., 2015; Fernandes & de las Mercedes Salas-Mellado, 2017) and mayonnaise (Fernandes & de las Mercedes Salas-Mellado, 2018), for the design of edible films (Capitani et al., 2016), and efforts have also recently been

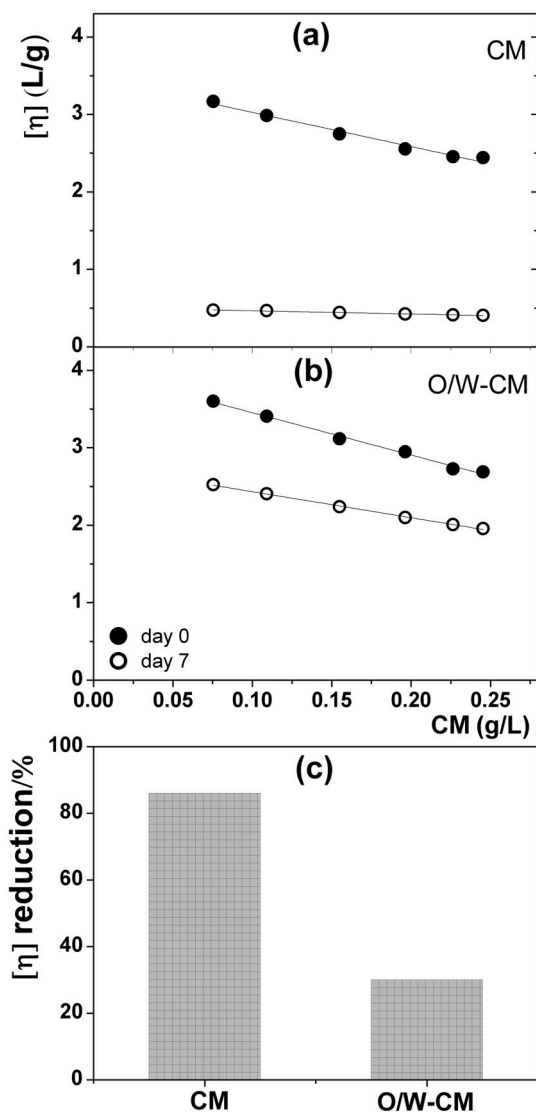


Fig. 4. Dependence of inherent viscosity for (a) CM suspension freshly prepared (full dots) and after seven days of storage (empty dots) and for O/W-CM (b) freshly prepared (full dots) and after seven days of storage. Dashed lines represent the linear fit of experimental points. (c) Reduction (%) of intrinsic viscosity, $[\eta]$, after 7 days of incubation at room temperature and gentle stirring.

made to understand at the nanoscale level the structure of the network provided by the mucilage fibers (Samateh et al., 2018). Moreover, also the aspect of the mucilage oxidation has never been considered before. We only found the paper by Hernández-Morales et al. (Hernández-Morales et al., 2019) where this aspect was positively used to carry out the green synthesis of metal-based nanoparticles.

On balance, our investigation can represent an interesting starting point for the realization of food-grade matrices in the food industry i.e. for the production of edible coatings and films or the use of chia mucilage as an ingredient in different food preparations.

Declaration of competing interest

The authors declare that there is no conflict of interest regarding the publication of this article. All co-authors agreed with the contents of the manuscript. I certify that the submission is original work and is not under review at any other publication.

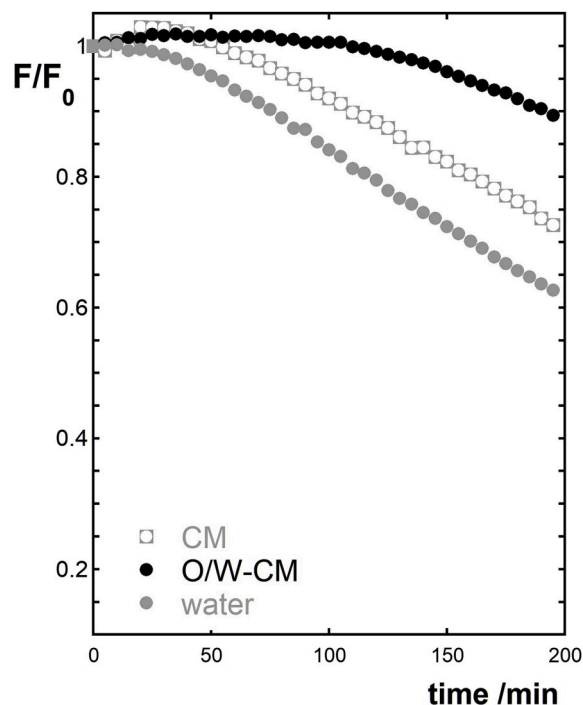


Fig. 5. Relative fluorescein emission decrease response as function of time in CM suspension, OW-CM and water.

CRediT authorship contribution statement

Francesca Cuomo: Conceptualization, Methodology, Software, Validation, Investigation, Data curation, Writing - original draft, Writing - review & editing, Supervision. **Silvio Iacovino:** Validation. **Maria Cristina Messina:** Validation. **Pasquale Sacco:** Investigation, Data curation. **Francesco Lopez:** Data curation, Writing - original draft, Writing - review & editing, Supervision.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.foodhyd.2020.105860>.

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