



Short Communication

Study on the possibility of developing food-grade hydrophobic bio-aerogels by using an oleogel template approach



Lara Manzocco, Federico Basso, Stella Plazzotta, Sonia Calligaris*

Department of Agricultural, Food, Environmental and Animal Sciences, University of Udine, Via Sondrio 2/A, 33100, Udine, Italy

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ABSTRACT

The feasibility of producing food-grade hydrophobic bio-aerogels by supercritical-carbon dioxide (SC-CO₂) extraction of oil from oleogels was investigated for the first time. Medium chain triglycerides (MCT) oil was gelled using ethylcellulose (EC) at increasing concentration (10, 15, 20% w/w) and grade (EC20, 45, 100), eventually in combination with fillers. Different SC-CO₂ oil extraction procedures were tested. The acquired results show that both oleogel formulation and extraction conditions can steer the EC scaffold structure. The increase in EC concentration and grade resulted in oleogels more structurally stable to SC-CO₂ extraction. The application of a pulsed extraction procedure allowed obtaining a low-density (0.39 g/cm³) EC scaffold presenting 60% oil. Addition of freeze dried lettuce powder improved macrostructure homogeneity. The obtained results lay the foundations for developing food-grade hydrophobic bio-aerogels, which are expected to present unique oil absorption and bioactive delivery features.

1. Introduction

Aerogels are porous materials presenting unique physical properties, including low density (0.0003–0.5 g/cm³), high surface area (50–1200 m²/g) and high porosity (70.0–99.8%) (Fricke and Tillotson, 1997). In the last years, they have attracted substantial interest also in the food sector, where they have been proposed as possible delivery systems for bioactive components (Selmer et al., 2015, 2019; Liang et al., 2017) and, more recently, for oil structuring (Plazzotta et al., 2019, 2020).

Up to now, bio-aerogels for food applications have been produced using hydrophilic biopolymers, including carbohydrates and proteins (Plazzotta et al., 2020; Ganesan et al., 2018). The typical production process of bio-aerogels involves three phases: 1) gelation of the biopolymer in water leading to a hydrogel; 2) water phase substitution by ethanol leading to an alcogel; 3) removal of ethanol by the application of a supercritical carbon dioxide (SC-CO₂) flow (Ganesan et al., 2018). The use of CO₂ is particularly advantageous being it food grade, non-flammable and eco-friendly.

Current bio-aerogels, being prevalently made of proteins and polysaccharides, present limited stability in moisture-rich environments, such

as most foods, in which they lose their structural integrity due to swelling and/or solubilization of the polymeric backbone (García-González et al., 2015). This limit might be overcome by developing hydrophobic bio-aerogels. Hydrophobicity could actually provide bio-aerogel with the unique advantage of being highly stable in the presence of water. A more hydrophobic aerogel backbone is also expected to boost the capability of aerogels to absorb lipids and lipophilic components, due to the higher surface affinity towards the absorbed molecules (Selmer et al., 2019). Nevertheless, aerogel hydrophobization requires cross-linking or silanization by using solvents and additives that are not food-grade. In this research, we proposed a novel approach to develop food-grade hydrophobic bio-aerogels based on the preparation of aerogels from oleogels rather than from hydrogels. In other words, the proposed strategy implies: (i) preparation of a food-grade oleogel, using a hydrophobic polymer able to structure edible oils; (ii) removal of oil from the oleogel by using SC-CO₂. To the best of our knowledge, the direct removal of oil from oleogels by SC-CO₂ has never been attempted, even if the oil removal from a water-in-oil emulsion was previously exploited to obtain bio-aerogels (Kleemann et al., 2018; García-González et al., 2012a). However, in this case the oil is substituted with ethanol by a solvent

Abbreviations: SC-CO₂, supercritical-carbon dioxide; MCT, medium chain triglycerides; EC, ethylcellulose; EC20, EC45, EC100, 20, 45 and 100 grade ethylcellulose; MG, monoglycerides; LP, lettuce powder; STD1, STD2, standard extraction procedures; SEQ, sequential extraction procedure; PSD, pulsed extraction procedure.

* Corresponding author.

E-mail addresses: lara.manzocco@uniud.it (L. Manzocco), basso.federico.1@spes.uniud.it (F. Basso), stella.plazzotta@uniud.it (S. Plazzotta), sonia.calligaris@uniud.it (S. Calligaris).

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exchange procedure before SC-CO₂ process. The lack of information is probably due to the difficulty of removing oil from oleogels without disrupting their polymeric network. To accomplish this difficult task, the oleogel polymeric network should be insoluble in SC-CO₂ and strong enough to support sample weight during oil removal. At the same time, the oil should have a molecular weight low enough to easily flow off the polymeric matrix during oil removal. Based on these considerations, the medium chain triglycerides (MCT) oil was structured with ethylcellulose (EC). MCT oil was selected being composed by fatty acids of 6–12 carbons, and thus particularly soluble in SC-CO₂ (Zuknik et al., 2016). This oil is also claimed to have interesting health benefits (Marten et al., 2006), so that its eventual residue in the aerogel would not be an issue. On the other hand, EC is a food-grade, non-SC-CO₂-soluble hydrophobic derivative of cellulose able to structure vegetable oils (Gravelle et al., 2018).

In this short note, we describe how hydrophobic EC-aerogels could be prepared by applying an oleogel template approach. To prove the concept, MCT oil was gelled using increasing concentrations of EC with different grade, added or not with fillers. The obtained oleogels were subjected to SC-CO₂ oil extraction under different methodological conditions. Dried scaffolds were then analyzed for visual structural integrity, mechanical strength, oil content, density and porosity.

2. Materials and methods

2.1. Reagents

Ethylcellulose 20 cP (EC20), 45 cP (EC45) and 100 cP (EC100) were obtained from Dow Ltd. (Calgary, AB, Canada), MCT oil (Cremer COOR MCT 60-40, fatty acid composition: C6:0 ≤ 0.5%, C8:0 55–66%, and C10:0 35–45%) from Cremer Oleo Division (Hamburg, Germany); liquid carbon dioxide (CO₂) (purity 99.995%) from Sapio (Monza, Italy), freeze-dried lettuce powder (LP) was produced by freeze-drying and grinding of *Iceberg* lettuce leaves (particle size (%): 200–250 μm: 92.8 ± 1, <200 μm: 8.0 ± 0.6 (Plazzotta et al., 2018a); composition (g/kg): humidity: 50 ± 1, total dietary fiber: 221 ± 2, proteins: 175 ± 8, ash: 42 ± 1, fat: 34 ± 1; carbohydrates: 479 ± 6 (Plazzotta et al., 2018b)), Myverol™ saturated monoglycerides (MG) (fatty acid composition: 1.4% C_{14:0}, 59.8% C_{16:0}, 38.8% C_{18:0}; melting point 68.05 ± 0.5 °C) from Kerry Ingredients and Flavour (Bristol, United Kingdom).

2.2. Preparation of oleogels

EC oleogels were prepared according to the methodology described by Davidovich-Pinhas, Gravelle, Barbut, & Marangoni (2015). EC20, EC45 and EC100 were dispersed in MCT oil at concentrations of 10, 15 and 20% w/w. The dispersions were then heated to 150 ± 10 °C upon gentle stirring for 15 min. The samples were set in a pre-heated oven at 150 °C for 180 min and then cooled to 25 °C. The obtained oleogels were then shaped as monoliths (20 ± 1.0 mm height and 15 ± 1.0 mm diameter). Additional samples were also prepared by adding 5% w/w MG or LP before heating the EC dispersions. Table 1 shows the sample composition.

2.3. Supercritical carbon dioxide extraction (SC-CO₂)

The SC-CO₂ extraction was performed directly on the EC oleogels in a plant developed at the Department of Agriculture, Food, Environmental and Animal Sciences of the University of Udine and previously described by Manzocco et al. (2017). Table 1 reports the oil extraction procedures applied for each sample, while Table 2 shows the process parameters for each SC-CO₂ considered treatment. Two standard extraction procedures (STD1 and STD2) were performed by placing the sample inside the 265 mL-volume stainless steel cylindrical reactor (Montur Srl, Codroipo, Italy) in which CO₂ was pressurized at different pressure and temperature. A continuous CO₂ flow was then set during the entire treatment

Table 1

Ethylcellulose grade and concentration, added fillers and concentration, and applied SC-CO₂ treatment procedure (STD standard, SEQ sequential, PSD pulsed) for each sample.

Sample	EC	EC concentration (% w/w)	Filler	Filler concentration (% w/w)	Process
A	20	10	–	–	STD1
B	20	15	–	–	STD1
C	20	20	–	–	STD1
D	45	20	–	–	STD1
E	100	20	–	–	STD1
F	100	20	–	–	STD2
G	100	20	–	–	SEQ
H	100	20	–	–	PSD
I	100	20	Monoglycerides	5	PSD
L	100	20	Lettuce powder	5	PSD

EC=Ethylcellulose, STD=Standard, SEQ=Sequential, PSD=Pulsed.

time (at 6.0 NL/min for 4.5 h). A slow decompression to atmospheric pressure was carried out at the same outlet flow value in 30 min. In the sequential extraction procedure (SEQ), CO₂ was pressurized under the same conditions of STD1 but the outlet flow was increased stepwise. The reactor was then depressurized to atmospheric pressure at 6.0 NL/min for 30 min. Finally, a pulsed procedure (PSD) was tested: the outlet flow was for 5 times shut down for 30 min. In all cases, at the end of the extraction, the absence of oil in the outlet flow was visually assessed. Analyses were performed one month after oil extraction to allow CO₂ complete removal.

2.4. Analytical determinations

2.4.1. Image acquisition

Images (72 dpi vertical and horizontal resolution) were acquired using an image acquisition cabinet (Immagini & Computer, Bareggio, Italy) equipped with a digital camera (EOS 550D, Canon, Milano, Italy) placed on an adjustable stand positioned 45 cm above a black cardboard base.

2.4.2. Mechanical strength

Mechanical strength was measured by uniaxial compression using an Instron 4301 (Instron Ltd., High Wycombe, UK). The instrumental settings and operations were accomplished using the software Automated Materials Testing System (version 5, Series IX, Instron Ltd., High 1 Wycombe, UK). Samples were tested using a 6.2 mm diameter cylindrical probe mounted on a 100 N compression head at a 25 mm/min crosshead speed. Force-distance curves were obtained from the compression tests and mechanical strength was taken as the Maximum Force (N) required to penetrate the sample for 5 mm.

2.4.3. Residual oil content

The oil content in aerogel was evaluated by weight difference between the oleogel before and after SC-CO₂ extraction.

2.4.4. Density

The density of the samples after oil removal was calculated dividing their weight by their volume. The latter was calculated based on sample height and diameter, assuming a cylindrical shape. Height and diameter of the monoliths were measured with a Metrica monobloc precision venier caliper (Metrica S.p.A., San Donato M.se MI Italy).

2.4.5. Porosity

The porosity of the samples after oil removal was estimated following equation (1).

$$Porosity (\%) = \frac{V_{eo} + \Delta V}{V_f} \times 100 \quad (1)$$

Table 2
SC-CO₂ oil extraction conditions applied of EC oleogels.

Process	Pressure (MPa)	Temperature (°C)	Flow	Process step	CO ₂ Flow (NL/min)	Time (h)	Total extraction time (h)
STD1	13.0 ± 1.0	45.0 ± 2.0	Continuous	1	6.0 ± 1.0	4.5	5.0
				2	Depressurization	0.5	
STD2	18.0 ± 1.0	60.0 ± 4.0	Continuous	1	6.0 ± 1.0	3.5	4.0
				2	Depressurization	0.5	
SEQ	13.0 ± 1.0	45.0 ± 2.0	Continuous	1	3.5 ± 0.5	3.0	8.5
				2	5.0 ± 0.5	4.0	
				3	6.0 ± 0.5	1.0	
				4	Depressurization	0.5	
PSD	18.0 ± 1.0	60.0 ± 4.0	Pulsed	1	3.0 ± 1.0	1.0	7.5
				2	0	0.5	
				3	3.0 ± 1.0	1.0	
				4	0	0.5	
				5	3.0 ± 1.0	1.0	
				6	0	0.5	
				7	3.0 ± 1.0	1.0	
				8	0	0.5	
				9	3.0 ± 1.0	1.0	
				10	Depressurization	0.5	

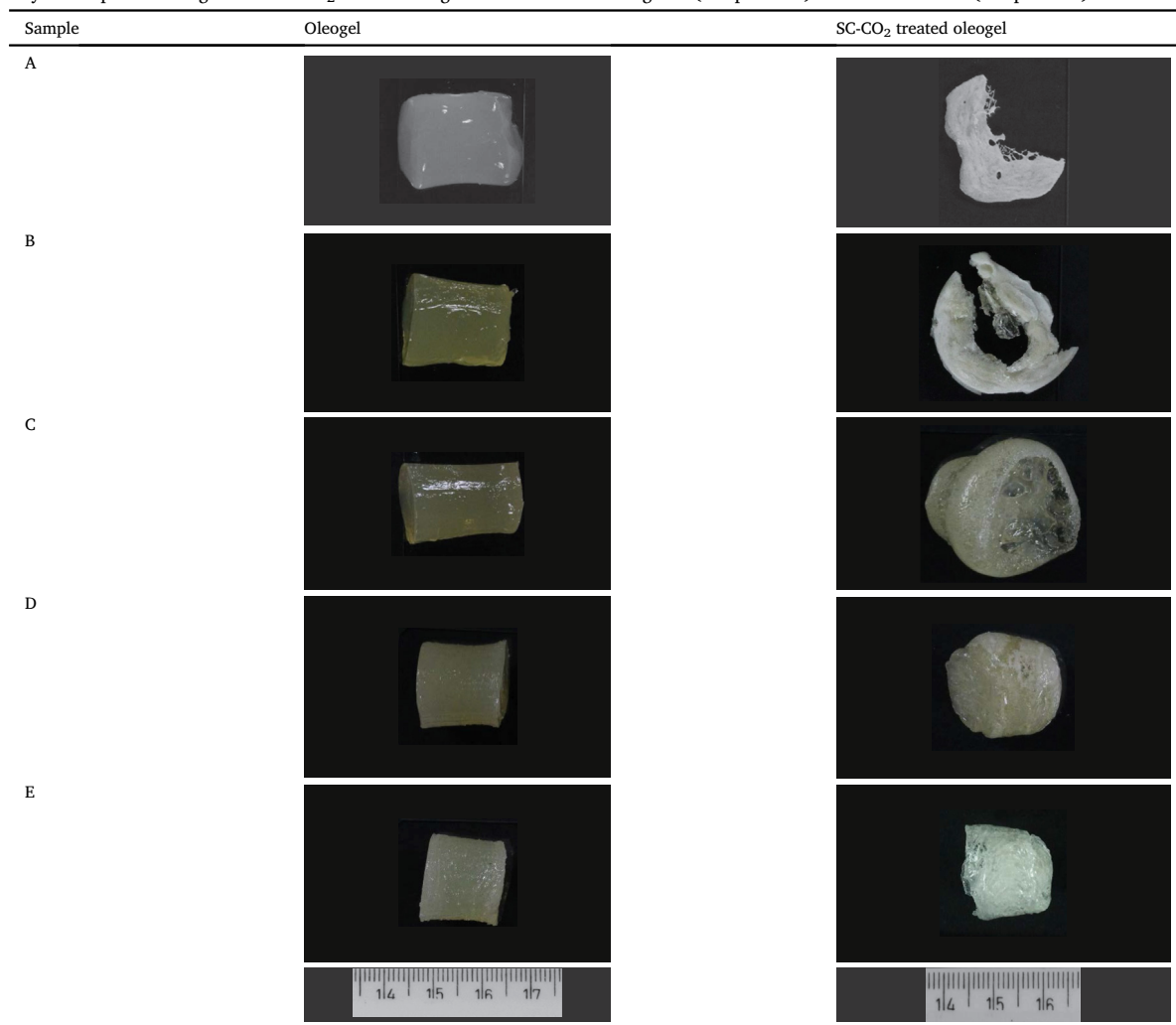
STD=Standard, SEQ=Sequential, PSD=Pulsed.

where V_{eo} (mL) is the volume of the extracted oil, which was calculated based on a MCT density of 0.92 g/cm³, ΔV (mL) is the volume variation before and after SC-CO₂ process and V_f (mL) is the volume of the sample after performing the extraction.

2.4.6. Data analysis

Sample preparation was performed in triplicate within the same experiment and all determinations were expressed as the mean ± standard error. Statistical analysis was performed using one-way analysis of

Table 3
Physical aspect of oleogels and SC-CO₂ treated oleogels as a function of EC grade (samples A-C) and concentration (samples C-E).



variance (ANOVA), followed by Tukey's Honest Significant Differences (HSD) test or *t*-test ($p < 0.05$).

3. Results and discussion

3.1. Effect of EC concentration

To prove the concept of the preparation of hydrophobic bio-aerogels by SC-CO₂ oil removal from EC oleogels, oleogels were initially prepared using EC20 at increasing concentration from 10 to 20% (w/w). The increase in EC concentration (samples A-C) resulted in oleogels with a more defined cylindrical shape (Table 3) and higher mechanical strength (Table 4). This result was expected and due to the higher content of EC, which allowed the formation of a more structured oleogel scaffold. Moreover, as the EC concentration increased the visible oil separation decreased. The obtained oleogels were subjected to the SC-CO₂ process to remove the oil. It should be noted that the conventional solvent exchange procedure consisting in substituting the solvent with ethanol before SC-CO₂ extraction was not applicable in this case due to the high solubility of EC in ethanol (Wasilewska and Winnicka, 2019). Preliminary trials demonstrated the EC network dissolution in ethanol.

Upon SC-CO₂ processing, the increase of oleogel mechanical strength was associated to a higher ability to withstand oil removal. In fact, the samples containing 10 and 15% EC (samples A and B) completely lost their shape during oil removal (Table 3). By contrast, sample C (20% EC) better maintained its original shape upon oil extraction.

After extraction, the overall oil content of the sample was about 60% (Table 4). Such result can be attributed to the oil affinity to EC, which interacts with oil mainly by hydrogen bonding with the triglycerides polar groups and with the polar species eventually present in the oil, but also by minor hydrophobic interactions with the triglycerides acyl chains (Gravelle et al., 2015). Being MCT extremely non-polar, it is expected to interact with EC chains primarily by hydrophobic interactions, which, however, are easily cleaved in favour of the interactions with SC-CO₂. Therefore, oil retention in the EC scaffold is probably mainly due to the collapse of the EC network upon solvent exchange from oil to SC-CO₂. However, further mechanisms could contribute to collapse. Although not specifically reported for oleogels, collapse of polymeric gels containing solvents other than oil (e.g. ethanol or acetone) has been reported in the literature (García-González et al., 2012b). During SC-CO₂ extraction, two main steps can be identified. In the first step, SC-CO₂ dissolves in the pore liquid, resulting in a fluid mixture composed by the gel liquid phase (ethanol, acetone, or oil) and CO₂. The liquid volume of such mixture increases with the SC-CO₂ concentration, until the maximum gel solvent holding capacity is reached. At this point, the liquid excess volume starts to leak, and extraction occurs primarily by convection due to the SC-CO₂ flow. As the process continues, a second extraction phase consisting on supercritical transition can be expected. During this second phase, the

Table 4

Mechanical strength of oleogels and SC-CO₂ treated oleogels as a function of EC grade (samples A-C) and concentration (samples C-E). Oil content and density of deoiled oleogels are also shown.

Sample	Oleogel		SC-CO ₂ treated oleogel	
	Mechanical strength (N)	Mechanical strength (N)	Oil content (% w/w)	Density (g/cm ³)
A	0.15 ± 0.02 ^d	N.D.	N.D.	N.D.
B	0.42 ± 0.03 ^d	N.D.	N.D.	N.D.
C	3.48 ± 0.50 ^c	N.D.	57.46 ± 7.54 ^a	N.D.
D	7.53 ± 0.02 ^b	5.35 ± 0.74 ^b	68.39 ± 8.98 ^a	0.57 ± 0.01 ^a
E	13.18 ± 0.69 ^a	7.70 ± 0.25 ^a	64.36 ± 6.34 ^a	0.55 ± 0.03 ^a

^{a,b,c,d} Different letters in the same column indicate significantly different means ($p < 0.05$).

N.D. Not determined.

extraction of the liquid phase is expected to be controlled by diffusion of the supercritical fluid mixture out the network pores, thus occurring much slower than during the first step (García-González et al., 2012b). In our experiment, fast oil removal from the EC network surface during the first step might have led to a sudden decrease of polymer-solvent interactions and a concomitant increase of polymer-polymer ones, accounting for a local structural collapse. It is likely that this surface layer acted as a barrier against the diffusion of SC-CO₂ through the gels structure, hindering the occurrence of the second extraction step and thus further oil removal.

3.2. Effect of EC grade

To reduce collapse during oil extraction, samples were prepared by changing the EC grade used for oleogel preparation (samples D and E). The increase in EC grade resulted in oleogels characterized by well-defined cylindrical shape, a reduced glossiness and with mechanical strength that increased with the EC grade (Table 4). These properties positively affected the oleogels ability to withstand oil removal and both scaffolds showed a density value in the typical range of aerogels (0.0003–0.5 g/cm³) (Fricke and Tillotson, 1997). The porosity of these samples after SC-CO₂ extraction resulted around 36% (sample D) and 64% (sample E). Although such values are lower than those reported for aerogels (70.0–99.8%) (Fricke and Tillotson, 1997), they suggest that the increase of EC grade greatly affects sample porosity. Although aerogels prepared with EC45 and EC100 (D and E) better maintained the initial shape as compared to EC20 (sample C), the collapse was still evident, leading to a residual oil content around 60% in all samples (Table 4).




3.3. Effect of oil extraction conditions

Based on the acquired results, the sample prepared with 20% EC100 was selected to tune SC-CO₂ treatment conditions. Since oil solubility could be a determining step for oil removal, both pressure and temperature were modified (STD2, Table 2). As shown in Table 5, the SC-CO₂ treated sample (F) resulted comparable to the one obtained at lower pressure and temperature (sample E, Table 3). An additional attempt was thus performed by gradually increasing the CO₂ flow from 3.5 to 6.0 L/min during oil removal (SEQ, Table 2 and sample G in Table 5). Also in this case, the obtained sample resulted similar to the previous one (sample F in Table 5). Accordingly, both sample F and G showed porosity values around 63%, comparable to that of sample E. A further trial was performed by decreasing the flow rate at 3.0 NL/min, which is in the range typically applied for production of polysaccharide-based aerogel from alcogels (García-González et al., 2012a). In addition, four cyclic interruptions of this flow rate (PSD, Table 2 and sample H in Table 5) were applied since equilibration of oil concentration during extraction is expected to allow progressive adaptation of the polymeric network to the decreasing solvent content (Manzocco et al., 2017). Although the obtained sample still showed the occurrence of collapse and an estimate porosity not significantly lower than that of sample F and G, it presented a density lower than that of the other samples, supporting the efficacy of PSD procedure in preventing network shrinkage. This result also underlines the critical role of flow rate and its application modality on the control of material structure during extraction.

3.4. Effect of fillers

The effect of the addition of two different fillers on EC SC-CO₂ treated samples was finally tested. In particular, monoglycerides (MG) were employed due to their ability of interacting with EC. It has been recently demonstrated that MG modified the rheological properties of EC oleogels by increasing EC solubility and enabling the hydrogen bonding among EC and hydroxyl groups of MG (Rodríguez-Hernández et al., 2021). Moreover, since MG crystallizes in MCT oil forming a gel network entrapping oil, the presence of this additional network beside that of EC could

Table 5Physical aspect, mechanical strength, oil content and density of SC-CO₂ treated oleogels made of 20% (w/w) EC100 subjected to different SC-CO₂ treatment conditions.

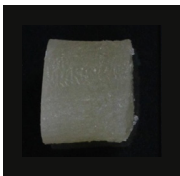

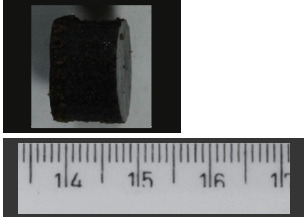

SC-CO ₂ treated samples	Images	Mechanical strength (N)	Oil content (% w/w)	Density (g/cm ³)
F		7.59 ± 2.31 ^b	63.40 ± 8.67 ^a	0.79 ± 0.01 ^a
G		8.64 ± 1.85 ^b	63.86 ± 9.06 ^a	0.43 ± 0.01 ^b
H		15.14 ± 1.36 ^a	60.63 ± 6.62 ^a	0.39 ± 0.01 ^c

^{a,b,c} Different letters in the same column indicate significantly different means ($p < 0.05$).

further reinforce the oleogel network (Fayaz et al., 2020). In agreement with these considerations, in our experiments we noted an increase of the oleogel mechanical strength as a consequence of MG addition (Table 6). On the other hand, lettuce powder (LP) was used due to its proven capability to prevent the physical collapse of other polymeric networks during supercritical-CO₂ drying (Plazzotta et al., 2018a, 2019). The LP sample (sample L, Table 6) showed a peculiar dark colour, due to partial carbonization of the vegetable powder during EC solubilization in oil at high temperature (150 °C). Interestingly, the LP oleogel mechanical strength resulted much higher than that of all the other samples. This effect could be attributed to the capacity of the LP components, such as cellulose, hemicellulose and proteins, to reinforce the oleogel network by strong self-association in lipophilic environment, leading to the formation of amorphous or partially crystalline supermolecular structures. In addition, it is not excluded that LP components could reinforce the oleogel network by establishing interactions with EC chains. Even if more studies should be done on this aspect, it is likely that these interactions might be initiated during the heating phase of the oleogel preparation

and further strengthened during cooling in a hydrophobic environment. Moreover, LP could also directly improve oleogels structuration by filling the polymeric network voids. Upon oil extraction, both MG and LP aerogels still presented about 60% oil content and a porosity value of 50 and 44%, respectively, which resulted lower than that of the filler-free sample (H) (Table 6). Moreover, the MG sample (I) showed a number of small white dots, suggesting that recrystallization of MG occurred following its partial solubilization during SC-CO₂ extraction at 60 °C. By contrast, the LP sample (L) not only showed a perfectly retained shape but also a much more homogeneous macrostructure, with significantly lower collapse. The visual evaluation of the sample revealed much less difference among the outer part and the sample core in comparison to the other samples. This observation could confirm the LP ability to reinforce the oleogel structure. Interestingly, oil extraction strongly reduced sample mechanical strength, which resulted comparable to that of the aerogel without fillers (sample H, Table 5). These results are in agreement with literature findings, showing that LP filler prevents the structural collapse of κ-carrageenan alcogels, leading to aerated aerogels (Plazzotta et al.,

Table 6Physical aspect of oleogels and SC-CO₂ treated oleogels made of 20% (w/w) EC100 added with 5% monoglycerides (sample I) or lettuce powder (sample L) and deoiled according to the PSD procedure. Oil content and density of deoiled oleogels are also shown.

Sample	Oleogel		SC-CO ₂ treated oleogel			
	Images	Mechanical strength (N)	Images	Mechanical strength (N)	Oil content (% w/w)	Density (g/cm ³)
I		17.56 ± 0.13 ^b		9.98 ± 0.44 ^a	60.90 ± 5.26 ^a	0.70 ± 0.02 ^a
L		48.92 ± 2.85 ^a		15.47 ± 4.65 ^a	57.36 ± 5.78 ^a	0.52 ± 0.04 ^b

^{a,b} Different letters in the same column indicate significantly different means ($p < 0.05$).

2019). In particular, the high mechanical strength of the sample is due to the counterbalancing effect of network maintenance and increased solid content. It can be concluded that, although the obtained systems do not properly fall under the classical bio-aerogel definition due to the high final oil content, their food-grade nature, low density, and strictly hydrophobic nature are expected to open new possibilities in the development of innovative food ingredients presenting unique oil absorption and bioactive delivery features.

4. Conclusions

Although just at the phase of proof of concept, oil removal from oleogels was demonstrated to be a promising strategy to develop food-grade hydrophobic bio-aerogels. Acquired results show that both oleogel formulation and extraction conditions affect the scaffold structure. Even if structure stability was improved by steering polymer concentration and size, complete oil removal was hindered by structural collapse.

A number of questions requires being answered before materials, having the desired structure and functionality, are achieved and characterized. What are the oleogels, among the many previously described, which could be turned into aerogels? What is the hydrophobicity of these new materials? What food-grade processes/ingredients could be used to modulate their hydrophobicity? How can oil be completely removed from the polymer network while preventing as much as possible its structural collapse? The latter is certainly the most challenging questions. Extracting oil by supercritical-carbon dioxide, rather than other solvents, is probably a good starting point. On this aspect, it is interesting to note that the process could be considered sustainable, since oil recovery could be obtained by passing the SC-CO₂-oil mixture through a specifically designed recovery device. Nevertheless, such an objective can be only accomplished by understanding, and properly managing, the mechanisms underlying oil retention within the polymer network. Answering these questions is a hard task, but the availability of food materials with very low density, and strictly hydrophobic nature, is expected to open new possibilities in the development of innovative ingredients presenting unique oil absorption and bioactive delivery features.

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CRedit authorship contribution statement

Lara Manzocco: conceived the original idea. **Federico Basso:** Formal analysis, Writing – original draft. **Stella Plazzotta:** Formal analysis, Writing – original draft. **Sonia Calligaris:** Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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