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Cold-Pressed Grape Seed Oil Encapsulation Using a Submerged-Nozzle Dispersion Encapsulation Process

Steva M. Lević [1,*], Vladislav A. Rac [1], Vesna M. Rakić [1], Ana S. Salević-Jelić [1], Marina R. Hovjecki [1], Marko V. Malićanin [2], Biljana B. Rabrenović [1], Mališa P. Antić [1] and Viktor A. Nedović [1]

[1] Faculty of Agriculture, University of Belgrade, Nemanjina 6, 11080 Belgrade, Serbia; vladarac@agrif.bg.ac.rs (V.A.R.); vesna.rakic@agrif.bg.ac.rs (V.M.R.); ana.salevic@agrif.bg.ac.rs (A.S.S.-J.); marina.hovjecki@agrif.bg.ac.rs (M.R.H.); biljanar@agrif.bg.ac.rs (B.B.R.); mantic@agrif.bg.ac.rs (M.P.A.); vnedovic@agrif.bg.ac.rs (V.A.N.)

[2] Faculty of Agriculture Kruševac, University of Niš, Kosančićeva 4, 37000 Kruševac, Serbia; malicanin.marko@ni.ac.rs

* Correspondence: slevic@agrif.bg.ac.rs

Abstract: Cold-pressed grape seed oil contains many bioactive compounds and its production enables the valorization of grape seeds as a by-product of winemaking. However, the incorporation of oils into food is associated with losses of active compounds and incompatibilities with the complex food matrix. Encapsulation is considered a process that could overcome these obstacles and enable a more diverse use of plant oils in the food sector. In this study, we developed an improved encapsulation process, submerged-nozzle dispersion (SND). SND aims at the encapsulation of active ingredients using highly viscous carrier materials. We used SND for the encapsulation of cold-pressed grape seed oil in alginate. The alginate stabilized the emulsions and provided stable conditions for the formation of encapsulates. The dried encapsulates were in the form of elongated particles with an average width below 150 µm; the oil content in encapsulates reached above 80% and encapsulation efficiency was up to 90%, depending on the formulation. Encapsulates exhibited satisfactory mechanical properties, suggesting they could mix well with other food ingredients. The SND encapsulation process developed in this study could be successfully applied to the encapsulation and protection of cold-pressed plant oils to be used as a source of valuable nutrients in foods.

Keywords: grape seed oil; encapsulation; alginate; submerged-nozzle dispersion

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1. Introduction

The consumption of foods rich in certain bioactive substances has become very popular in recent decades. This trend is leading to changes in the food industry through the introduction of new processes and resources to create new products rich in health-promoting ingredients. Plant oils obtained by cold pressing are considered a source of important compounds that are characterized by their beneficial effects on the biological processes in the human body, as the extraction conditions are mild and no additional chemical treatments are required [1].

Grape seed oil is a valuable source of numerous important health-promoting components such as unsaturated fatty acids, polyphenols and tocopherols. One of the main advantages of grape seed oil is that this oil is extracted from food waste, i.e., grape seeds left over after winemaking and grape processing. In addition, the new extraction processes can significantly influence the chemical composition and quality of grape seed oil and could be considered as an overall alternative to conventional solvent extraction [2].

The protection of food and certain food ingredients from damage due to unfavorable processing and storage conditions is one of the main challenges. Therefore, additional strategies for the preservation of sensitive food ingredients are needed. Encapsulation could be considered a suitable approach for the protection of various food ingredients, especially

plant oils, from the negative influence of high processing temperatures, light, oxygen, etc. The protection of the target substance by encapsulation is based on the formation of a layer or layers of selected materials, i.e., carrier materials, which should form a barrier against unfavorable environmental conditions [3]. In addition, a controlled release of the encapsulated compound under certain conditions could be achieved by selecting specific carriers (e.g., thermal release or release under certain conditions such as those in the human gastrointestinal system) [4,5]. The primary methods used to encapsulate plant oils are spray drying, freeze drying, complex coacervation, ionic gelation, in situ polymerization, etc. Although spray drying and freeze drying are widely used in the food sector, the oil capsules produced by these methods usually have a low oil content, while the number of carriers is limited, and operating costs can be high. Ionic gelation is based on the entrapment of the active ingredient in a polymer network (e.g., sodium alginate in reaction with Ca^{2+} cations), resulting in an insoluble structure with a relatively high oil content. In addition, the formation of these encapsulates can be controlled by applying suitable extrusion techniques and by optimizing the process conditions [6].

Alginate has been widely used for the encapsulation of active ingredients by extrusion encapsulation processes. Heck et al. [7] used alginate and an air dispersion to produce chia oil–alginate encapsulates that were incorporated into burgers.

The encapsulation process could require a change in the composition of the carrier material. In this context, alginate could be mixed with other components such as gelatin [8] or solids such as starch [9] before extrusion.

Besides the good properties of alginate and its gels intended for food applications, some limitations of alginate/plant oil encapsulation and encapsulation processes should be pointed out. The dispersion and formation of encapsulates with a suitable size and shape could be a challenge. This is because alginate-based emulsions exhibit changes in viscosity compared to the initial alginate solution [5]. Therefore, the conditions of the encapsulation process must be optimized each time the composition of the starting emulsion is changed. In addition, the oil changes the specific gravity of the emulsion and, consequently, the properties of the droplets and the resulting encapsulates [10].

When preparing relatively large amounts of emulsion with the encapsulation system shown in Figure 1a, we found in our preliminary study that the droplets could not overcome the interface between air and liquid (i.e., the gelling solution), so they floated on the surface of the gelling bath. As encapsulation proceeded, the newly formed droplets accumulated on the surface of the gelling bath and formed an almost compact layer that hindered the gelling process and the formation of encapsulates. Even with vigorous mixing of the gelling solution, some materials still clumped together. To overcome these difficulties, we propose in this study an encapsulation process with submerged-nozzle dispersion (SND).

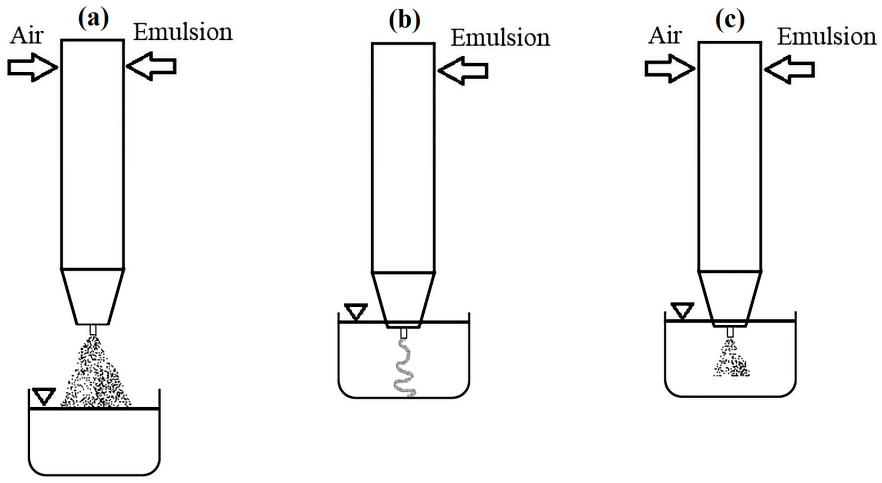

Figure 1. Schematic illustrations of dispersion encapsulation systems: (**a**) air dispersion; (**b**) wet spinning; (**c**) SND encapsulation process.

The idea behind the SND encapsulation process is to immerse the air dispersion nozzle (similar to wet spinning, Figure 1b) under the surface of the gelling bath (Figure 1c). In this way, the interface between the air and the gelling solution is no longer an obstacle to the formation of encapsulates. Compressed air is used as a dispersing fluid, which interrupts the flow of the encapsulated liquid (i.e., the alginate/oil emulsion) and causes dispersion and encapsulate formation. The SND process is similar to immersed microfluidic spinning, which is proposed for the production of alginate fibers primarily intended for tissue engineering [11]. Although both processes extrude the material below the surface of the gelling bath, the SND process uses compressed air to break the continuous fiber into smaller particles. According to Lević et al. [5], alginate reacts rapidly with Ca^{2+} ions (the most commonly used gelling agent) to form a stable matrix that entraps the active ingredient. The process requires a certain amount of time to ensure complete gelation of the alginate and the formation of Ca-alginate. The encapsulates are then removed from the gelling solution and washed with water. To preserve the active ingredient and ensure safe storage and longer shelf life, the encapsulates are usually dried [5]. As can be seen, the SND encapsulation process is complementary to other extrusion processes in terms of preparation and finalization steps. Therefore, the implementation of the SND encapsulation process into existing processes could be achieved without significant investment.

In this study, the alginate-based encapsulates containing cold-pressed grape seed oil were prepared using a newly developed encapsulation process (submerged-nozzle dispersion, SND). We tested the basic parameters of the grape seed oil–alginate emulsions and evaluated their influence on the properties of the encapsulates. This included evaluating the oil content and encapsulation efficiency as well as examining the morphology of the encapsulates using light and scanning electron microscopy. The color properties of the encapsulates and their texture were analyzed as important parameters for the use of the encapsulates in food.

2. Materials and Methods

2.1. Materials

Sodium alginate and calcium chloride dihydrate were supplied from Carl Roth (Karlsruhe, Germany). The grape seeds of the grape variety Pinot Noir were collected from the Zmajevac winery (Serbia), in the 2023 season. The cold-pressed grape seed oil was obtained using a UM200 cold-press oil machine (Ulimac Machine, Ankara, Türkiye). The grape seed moisture content was 8% w/w and the temperature of the obtained oil at the exit from the oil press was in the range of 44–46 °C. Solids that remain in the oil were removed by centrifuge (U-320, Boeco, Hamburg, Germany) at 5000 rpm for 5 min. Other used chemicals were of analytical reagent grade.

2.2. The Submerged-Nozzle Dispersion (SND) Encapsulation Process

Encapsulation of the cold-pressed grape seed oil was realized using a submerged-nozzle dispersion (SND) encapsulation process developed for this purpose (Figure 1c). The SND encapsulation process is based on the dispersion of oil–alginate emulsion into a calcium chloride gelling solution. Sodium alginate solution (1.5% w/w) was homogenized with 2.5, 5, and 10% (w/w) of cold-pressed grape seed oil (Table 1) using a mechanical homogenizer Ultra-Turrax® T25 (Janke and Kunkel Ika-Labortechnik, Staufen, Germany), at 10,000 rpm for 5 min. Immediately after preparation, the emulsion was processed using an SND system (Figure 1). The emulsion was transferred to the SND system by a peristaltic pump (MasterFlex® model No. 7523-27, Cole-Parmer, Barrington, IL, USA) at flow rate of 10 mL/min. Calcium chloride solution (1.5% w/w, at room temperature) was used for the gelling of the dispersed alginate/oil emulsion. An SND nozzle (1 mm inner diameter) was submerged into the calcium chloride solution at about 1 cm from the surface and approximately 3 cm from the bottom. During dispersion and an additional gelling period of 1 h, the obtained encapsulates were stirred using a magnetic stirrer. Compressed air was used for the dispersion of the emulsion into the gelling solution at 6 bar while the air

flow rate was set at 355 L/h. After the gelling period, encapsulates were collected from the gelling solution using a vacuum filtration and paper filter, followed by several washings with distilled water to remove the remaining calcium chloride, combined with vacuum filtration (by water-jet pump). Prior to freeze drying, the encapsulates were mixed with distilled water (at a mass ratio of wet encapsulates—water of 0.8:1) to prevent encapsulate agglomeration, followed by freezing at −80 °C for 24 h in plastic Petri dishes. Control encapsulates were freeze dried without the addition of water in order to evaluate the effects of water as a drying aid/dispersive agent. The freeze drying was performed using an Alpha 1-4 LSC plus laboratory freeze dryer (Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany) under the following conditions: −20 °C and a vacuum of 0.1 mbar for 10 h; −10 °C and a vacuum of 0.1 mbar for 5 h; 0 °C and a vacuum of 0.1 mbar for 5 h; 25 °C and a vacuum of 0.05 mbar for 4 h. The dried samples were stored in closed vials at ~4 °C.

Table 1. Sample formulations and properties of emulsions used for encapsulation of grape seed oil by SND encapsulation process.

Sample (Emulsions)	Grape Seed Oil Content in Emulsion (% *w*/*w*)	Emulsion Stability (%)	Comment
SND 2.5	2.5	~100	The emulsion had a white color
SND 5	5	~100	The emulsion had a white color
SND 10	10	~100	The emulsion had a pale cream/yellow color

2.3. Emulsions' Stability

Emulsion stability was evaluated according to the method previously described by Lević et al. [5]. Immediately after preparation, the emulsions (about 50 mL) were transferred into graduated cylinders and left for 1 h. The emulsion stability was expressed in % as a ratio of emulsion volume that is still stable after 1 h and the initial emulsion volume. The results are based on three independent measurements.

2.4. Light Microscopy

The morphological properties of wet encapsulates were analyzed using a Leica DMLS light microscope (Leica, Wetzlar, Germany) equipped with a Leica-DC 300 camera (Leica, Wetzlar, Germany) and using the Leica-IM 1000 software (Leica, Wetzlar, Germany).

2.5. Scanning Electron Microscopy (SEM)

The scanning electron microscopy analysis of the dried encapsulates was carried out using a JSM 6390LV scanning electron microscope (JEOL, Tokyo, Japan). The samples were prepared for SEM analysis by covering them with gold using a sputter coater model scd 005 (Baltec, Balzers, Liechtenstein).

2.6. Analysis of Water Content

The water content of the dried encapsulates was analyzed using the drying method at 105 °C. The samples were measured and dried in the oven until a constant weight was achieved. The moisture content was calculated from the weight loss between the initial and the dried sample [12].

2.7. Color Measurement

The color of the dried encapsulates was analyzed by a Chroma Meter CR-400 (Konica Minolta, Tokyo, Japan), using a D65 light source and an observer angle set at 2°. A standard white plate was used as a calibration standard. Color parameters were obtained based on 5 measurements.

2.8. Encapsulation Efficiency and Oil Content in Encapsulates

Encapsulation efficiency and oil content were analyzed after dissolution of encapsulates and oil extraction. Briefly, 2 g of dried encapsulates were dissolved in 40 mL of 3% (*w*/*v*) sodium citrate solution for 24 h on a magnetic stirrer at room temperature. For oil extraction, 15 mL of n-hexane was added, and extraction was carried out for 5 h under the same conditions as dissolution. Water–organic phase separation was performed using a centrifuge (U-320, Boeco, Hamburg, Germany) at 5000 rpm for 10 min. Separation of oil from n-hexane was performed at room temperature for 24 h followed by heating in a heating oven (UF 55, Memmert GmbH + Co.KG, Schwabach, Germany) at 40 °C until constant weight. The encapsulation efficiency (%) was calculated as a ratio between the mass of extracted oil and the initial mass of oil. Oil content (%) was calculated as a ratio between the mass of extracted oil and the mass of encapsulates.

2.9. Texture Analysis of Encapsulates

The hardness and stickiness of materials were analyzed by mechanical compression on the texture analyzer (Stable Micro Systems, TA.XT Plus, Godalming, UK) according to Xia et al. [13] with some modifications. Approximately 2 g of samples were measured in a stainless-steel cylindrical sample holder and placed on the heavy-duty platform on the flat insert with target centering rings. A cylinder probe with a diameter of 5 mm was used for analysis, the trigger force was set to 5 g, and the distance of probe penetration into the sample was 5 mm. The test speed of the probe was 1 mm/s. Three measurements were performed on each sample.

2.10. Statistical Analysis

Analyses were performed at least in triplicate and data are presented as mean ± standard deviation (unless otherwise stated), using PAST software (version 4.12b) [14]. One-way ANOVA was applied to determine significant differences between the physical parameters of encapsulates. The Tukey test, or the Kruskal–Wallis and Mann–Whitney tests, was used at $p < 0.05$ to identify significant differences among the samples.

3. Results

3.1. Properties of the Grape Seed Oil–Alginate Emulsions

Stable grape seed oil–alginate emulsions were prepared in the first step to produce oil-containing encapsulates using an SND encapsulation process. Our results showed that alginate stabilized the initial grape seed oil emulsions for a sufficient time to perform encapsulation and obtain stable encapsulates (Table 1). We did not observe phase separation or strong foaming during emulsion production.

3.2. Morphological Properties of the Grape Seed Oil–Alginate Encapsulates Obtained by the SND Encapsulation Process

The microscopic images of the wet encapsulates are shown in Figure 2a. The overall appearance of the encapsulates is the same regardless of the formulation. Under the conditions of the SND encapsulation process, the viscous oil–alginate emulsion is extruded into filaments. The gelling process of the alginate starts immediately upon contact with the gelling solution. However, the compressed air breaks up the filaments into irregularly elongated particles. The extruded particles continue the gelling process and the formation of insoluble calcium alginate, effectively trapping the oil. The process is complete when the volume ratio of the extruded emulsion to the gelling solution is 1:3. After the gelling phase (i.e., additional gelling process after extrusion in the same gelling solution), the capsules were washed with water and dried as previously described.

In general, dried encapsulates are preferable, as a low water content provides better conditions for the preservation of sensitive ingredients. Freeze drying was used as the drying method. After freeze drying, the encapsulates showed different morphological properties depending on the amount of water added before drying. After the encapsulates

were formed, vacuum filtration was performed to separate them from the gelling solution and the water used for washing. Vacuum filtration resulted in the formation of a thick filter cake. As can be seen in Figure 2(b1), the encapsulates that were dried without adding water before freezing (i.e., the preparation for freeze drying) were in the form of larger aggregates with a loose structure. The reason for this is probably the formation of a compact filter cake during filtration and separation of the encapsulates, which remain slightly compressed even after freeze drying. On the other hand, the encapsulates with added water (Figure 2(b2)) exhibited a looser structure and can generally be considered more suitable for incorporation into food matrices.

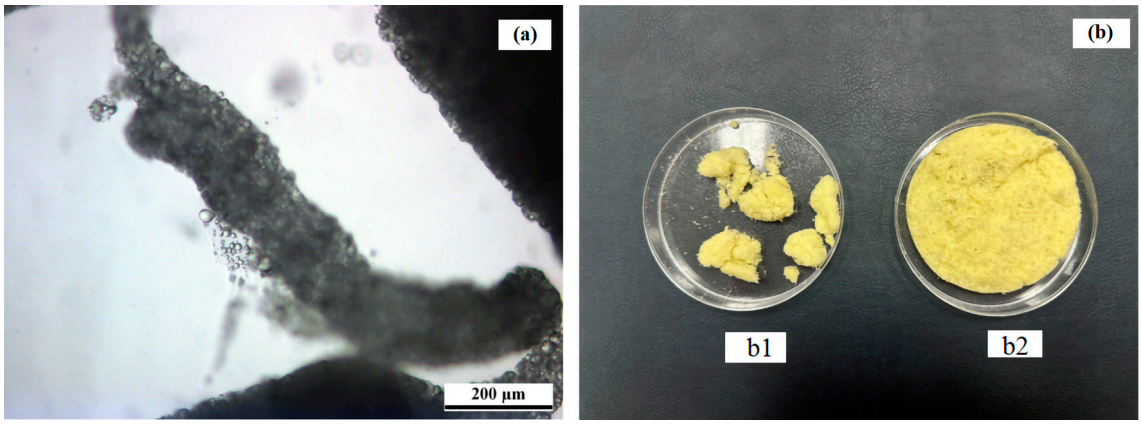

Figure 2. (**a**) Wet encapsulates observed by light microscopy; (**b**) the macroscopic appearance of freeze-dried encapsulates: (**b1**)—encapsulates dried without additional water, (**b2**)—encapsulates dried with added water.

3.3. Color Properties of the Grape Seed Oil–Alginate Encapsulates Obtained by the SND Encapsulation Process

As can be seen in Table 2, the values for L^* can be interpreted as high, which generally indicates light-colored samples. The green tone of the samples, which is defined by negative a^* values, is due to the green color of the cold-pressed grape seed oil. The yellow color component of the encapsulates was confirmed by the positive b^* values. The overall color properties of encapsulates could be defined as similar and influenced by oil content.

Table 2. Color properties of the grape seed oil–alginate encapsulates obtained by the SND encapsulation process.

Sample (Encapsulates)	L^*	a^*	b^*
SND 2.5	82.25 ± 0.5^{a}	-4.53 ± 0.04^{a}	22.81 ± 0.43^{a}
SND 5	79.28 ± 0.92^{b}	-5.71 ± 0.07^{b}	25.36 ± 0.93^{b}
SND 10	81.28 ± 0.93^{a}	-6.34 ± 0.09^{c}	25.99 ± 0.81^{b}

Different letters in superscript within the same column indicate significant differences among samples ($p < 0.05$).

3.4. Microstructural Properties of the Dried Grape Seed Oil–Alginate Encapsulates Produced by the SND Encapsulation Process

The SEM images (Figure 3) show that the dried encapsulates have elongated and irregular shapes, similar to the wet encapsulates. The encapsulate formulation SND 2.5 shows a more compact structure, while the shape of the encapsulates becomes more distorted with increasing oil content. The reason for this is most likely that the increased oil content (in the original emulsions) disturbed the water phase, i.e., the alginate, resulting in uneven formation of the calcium alginate network and distortion of the shape of the encapsulates. Although all encapsulate formulations showed a particle width of less than

150 µm, the size distribution is very broad, which emphasizes the complexity of encapsulate formation in the SND process. However, once formed, the encapsulates remained stable, which was confirmed by SEM analysis and analysis of oil content and encapsulation efficiency (see below). At higher magnifications (Figure 3), it was also found that a higher oil content affected the surface properties of the encapsulates, causing a rougher surface. After drying, the encapsulates shrank as the water evaporated and the oil droplets became more densely packed. This process is clearly visible on the surface of the encapsulates as small lumps.

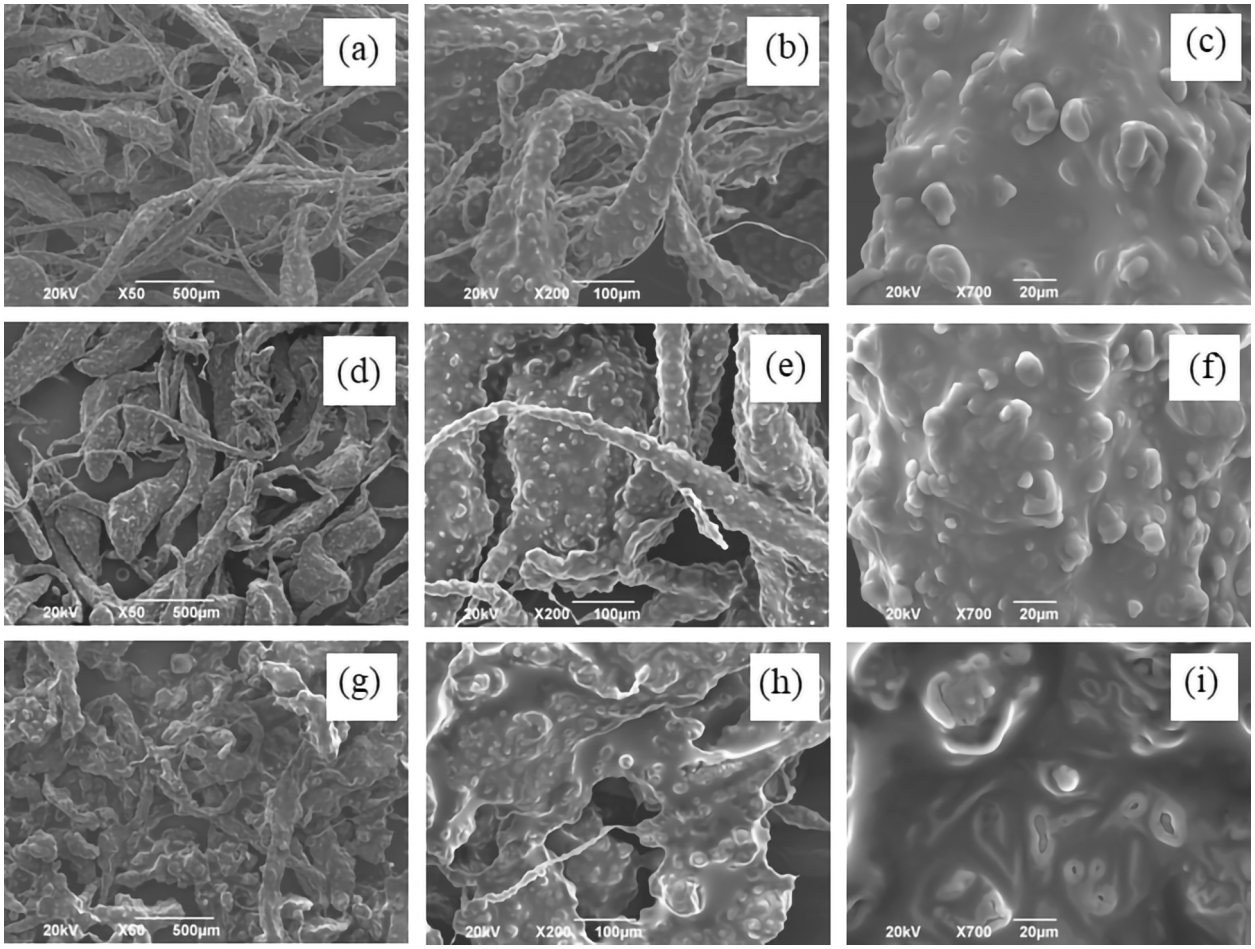

Figure 3. SEM images of dried encapsulates: SND 2.5 (2.5% *w*/*w* initial oil content) (**a**–**c**); SND 5 (5% *w*/*w* initial oil content) (**d**–**f**); SND 10 (10% *w*/*w* initial oil content) (**g**–**i**).

Based on data on the width of the dried encapsulates (Table 3), it was found that the dry encapsulate size was stabilized with added oil (i.e., oil is also a stabilizing filler).

Table 3. The main properties of dried encapsulates loaded with cold-pressed grape seed oil produced by the SND encapsulation process.

Sample (Encapsulates)	Dry Encapsulates Width (µm)	Moisture Content (%)	Encapsulation Efficiency (%)	Oil Content (%) *
SND 2.5	63.6 ± 49.7 ^{a}	5.6 ± 0.1 ^{a}	92.7 ± 1.8 ^{a}	55.5 ± 3.7
SND 5	116.7 ± 82.1 ^{b}	3.1 ± 0.3 ^{b}	89.4 ± 2.5 ^{a}	68.1 ± 3.1
SND10	145.1 ± 56.8 ^{c}	2.2 ± 0.2 ^{c}	88.7 ± 4.1 ^{a}	82.2 ± 2.4

* Values were not statistically compared due to initial differences in the oil content of each formulation. Different letters in superscript within the same column indicate significant differences among samples ($p < 0.05$).

3.5. Encapsulation Efficiency of the SND Encapsulation Process

The results of the encapsulation efficiency and the oil content for the encapsulates are shown in Table 3. It can be seen that the oil content of the encapsulates increases from ~55% to above 80%, with increasing oil content in the initial emulsions. This was achieved thanks to a high encapsulation efficiency of about 90%, which is very promising considering that no additional emulsifiers were used. Also, encapsulation efficiency slightly decreased with increasing oil content suggesting that alginate could sustain even higher oil concentrations.

3.6. The Hardness and Stickiness of the Encapsulates

During food processing, added encapsulated ingredients should remain in their original form, i.e., they should retain their mechanical stability. Therefore, the grape seed oil encapsulates produced using the SND encapsulation process were analyzed for their mechanical (i.e., textural) properties; the results are shown in Table 4.

Table 4. The hardness and stickiness of the grape seed oil encapsulates obtained by the SND encapsulation process.

Sample (Encapsulates)	Hardness (mN)	Stickiness (mN)
SND 2.5	2016 ± 240^{a}	-0.8 ± 0.3^{a}
SND 5	1780 ± 451^{a}	-1.2 ± 0.2^{a}
SND 10	402 ± 68^{b}	-2.1 ± 0.3^{b}

Different letters in superscript within the same column indicate significant differences among samples ($p < 0.05$).

The hardness of the encapsulates decreased with increasing oil content. These results are to be expected as the alginate network is weakened by the oil droplets, resulting in less resistance to the applied force. The stickiness of encapsulates was also affected by the ratio between the oil and alginate, i.e., it increased with increasing oil content.

4. Discussion

The application of encapsulated active food compounds in a food matrix is challenging as the encapsulates are associated with limitations, such as undesirable size, low encapsulation efficiency, incompatibility with the processing of the main food, etc. Therefore, the optimization and regulation of encapsulate properties is necessary to successfully incorporate them into food formulations [4]. Alginate is a suitable carrier material that simultaneously provides emulsion stability and forms a stable and compact matrix that separates the active ingredients from the environment [5]. To overcome the problems associated with the encapsulation of high-content plant oils, we have developed an SND process aimed at encapsulating oils in alginate or potentially other carriers with a similar gelling mechanism.

The fact that alginate is an emulsion stabilizer helped in the formulation of the encapsulates and simplified their production. Lević et al. [5] showed that alginate serves as an emulsifier and carrier in the formation of encapsulates with hydrophobic compounds. To further improve the properties of grape seed oil–alginate emulsions, the addition of emulsifiers prior to homogenization could result in emulsions with better/prolonged stability and even higher oil content. For example, whey protein isolate showed promising results in stabilizing the emulsion before mixing with alginate and encapsulate formation [15]. Nevertheless, in this study, even without an additional emulsifier, alginate–oil emulsions were successfully processed into stable encapsulates using the SND encapsulation process under optimized conditions.

The SND encapsulation process showed satisfactory performance in the production of encapsulates with different oil concentrations (Table 1). SND opens up possibilities for the encapsulation of various active ingredients and biocatalysts. Alginate is a well-established carrier system for the encapsulation of probiotic bacteria [8], bioactive compounds [16],

and food additives [5]. However, due to the high viscosity of alginate solutions, existing encapsulation methods could generate elongated particles [5]. Our SND encapsulation process also yields elongated forms of encapsulates (see below). Other highly viscous carrier materials could be used in the SND encapsulation process. Pectin, for example, which can also form gels through ionic gelation [17], is a promising carrier for the SND encapsulation process. Gelatin could also be mixed with alginate to produce more complex encapsulates that enable the controlled release of encapsulated active compound in the gastrointestinal tract [8].

The main limitations of the SND encapsulation process that were identified are related to the application of higher compressed air flow rates; the encapsulation efficiency decreased with increasing air flow rate. Namely, a higher air flow rate should provide smaller particles and thus encapsulates with a favorable size. However, during the optimization steps, we found that it is possible to reduce the size of the encapsulations, but with a lower encapsulation efficiency, i.e., a lower oil load. The reason for this is probably the release of oil from the emulsion as a result of the higher shear stress at higher airflows. Increased shear stress at the nozzle tip interrupts the emulsion flow, which leads to the release of oil droplets into the gelling solution and consequently reduces the encapsulation efficiency. Alginate is a polysaccharide without hydrophobic groups. Therefore, alginate is not considered a strong emulsifying agent. In this context, improving the emulsifying properties of alginate could be achieved by non-ionic surfactants [18]. For future studies, it would be interesting to investigate whether smaller encapsulates with high oil loads can be achieved with the SND encapsulation process using additional emulsifiers. However, in this study, the encapsulation conditions in the SND encapsulation process were optimized so that encapsulates with a high oil load and encapsulation efficiency could be produced without using additional emulsifiers.

An additional material is usually added before the encapsulated ingredient is freeze dried to form a protective barrier and ensure good mechanical properties of the powder. Materials such as maltodextrin and protein isolates have been widely used for freeze-drying encapsulation of plant oils [3]. The use of carbohydrates as drying aids could be challenging due to their chemical and nutritional properties. Namely, freeze-dried carbohydrate-based encapsulates usually exhibit significant hygroscopicity [19], which complicates their handling and application in food formulations. There is also growing concern about the increasing use of carbohydrates, especially processed carbohydrates such as maltodextrin, which can increase glycemic load, potentially negatively impacting consumer health [20]. Therefore, we used water as a drying aid (Figure 2) and dispersant to avoid the use of carbohydrates and produce encapsulates without additional calories, suitable for consumption by a wider population.

The color of encapsulates is a very important property for their use in foods, which could consequently affect the consumers' visual perception. In this regard, encapsulation can change the color of the active ingredient, and this phenomenon depends on the color of the active ingredient and the carrier material [12]. In this study, we used the three-dimensional CIE $L^*a^*b^*$ color space to evaluate the color of the encapsulates (Table 2). In the CIE $L^*a^*b^*$ color space, the L^* indicates the lightness of the sample (light = high L^* values or dark = low L^* values). Values for a^* in the positive direction define a red tone, while negative a^* values indicate a green tone. Positive values for b^* point to a yellow coloration and negative values indicate a blue coloration of the sample [21]. The changes in the values for the parameters a^* and b^* in the described direction and with increasing oil content show that the color of the encapsulates was primarily influenced by the color of the cold-pressed grape seed oil. The color of the SND encapsulates loaded with cold-pressed grape seed could be considered as suitable for mixing with various food matrices. Furthermore, the color properties of the encapsulates open up possibilities for the use of additional food colors (to be applied in encapsulate formulations) for the creation of the desirable visual properties of foods.

Dry encapsulates are more suitable for storage, and a low moisture content is generally considered beneficial to minimize undesirable chemical reactions or microbial spoilage. Therefore, moisture content should be kept as low as possible, especially for encapsulates containing sensitive compounds such as plant oils [22]. Freeze drying is a suitable method to remove most of the moisture from the organic material while retaining most of the structural properties of the sample. The results for the moisture content of the encapsulates (Table 3) show that this parameter depends on the oil content, i.e., the higher the oil content in the encapsulates, the lower the moisture content. Nevertheless, the low moisture content of SND encapsulates could be the basis for the development of encapsulated plant oils suitable for prolonged storage.

The morphology of cold-pressed grape seed oil encapsulates obtained by SND process were affected by addition of oil. Lević et al. [5] reported a similar influence of the hydrophobic agent (D-limonene) on the formation of Ca-alginate beads during electrostatic extrusion encapsulation. Shrinkage of alginate-based encapsulates occurs during drying and depends on the concentration of the alginate and the active ingredient. The presence of an active ingredient can maintain the shape and size of the dried encapsulates to a certain degree [5]. Córdoba et al. [9] showed that the addition of corn starch stabilized the spherical shape of the encapsulates, but additionally impaired the release of the active ingredients under simulated gastrointestinal conditions. The SND encapsulation process could be used for encapsulation of these complex encapsulation systems since it is not significantly affected by changes in carrier composition.

The encapsulates showed high encapsulation efficiency that is in agreement with the literature data on the encapsulation of oils within polysaccharide carriers cross-linked by ionotropic gelation. Heck et al. [7] reported an encapsulation efficiency of ~87% for chia oil–alginate encapsulates obtained by air dispersion. Menin et al. [17] showed that pectin-based encapsulates with flaxseed oil have a high encapsulation efficiency of up to 98%. The results for oil content and encapsulation efficiency obtained with the SND encapsulation process could be the basis for further development of new encapsulates with specific properties intended for food applications. A high oil content is one of the main requirements in the development of encapsulates, meaning that fewer encapsulates are needed to achieve the delivery of a specific quantity of encapsulated compound.

According to Schädle et al. [23], hardness and stickiness are also important for the sensory properties of food. It could be expected that the hardness of encapsulates could be controlled by changes in oil and alginate concentrations. Therefore, a more concentrated carrier solution will most likely provide better mechanical properties of the encapsulates. However, the viscosity of polymers such as alginate increases significantly with increasing concentration [5]. As we have already mentioned, viscous carrier solutions are difficult to process in standard extrusion encapsulation processes. Therefore, our SND encapsulation process could be used to produce more mechanically stable encapsulates. Furthermore, to improve the mechanical properties of alginate-based carriers, different materials could be added before encapsulation. Park et al. [24] used cellulose nanofibrils to reinforce alginate fibers produced by wet spinning. However, this reinforcement depended on the concentration of cellulose nanofibrils, with mechanical properties deteriorating after a certain concentration of cellulose was reached. The selection of suitable carrier material to reinforce the base carrier must therefore be optimized according to the specific requirements. On the other hand, the hardness of the encapsulates should be in the ranges that match the acceptable hardness of food and the expected mouthfeel as an important sensory property of food. For example, the hardness of the SND 10 encapsulates (with higher oil content) was around 400 mN, which is far below the values expected for the hardness of meat and meat-substituted products [25]. Even encapsulates with lower oil content exhibited a hardness that could allow their use in meat products to improve their nutritional properties.

Stickiness is generally considered to be a property of materials, especially powders, which becomes more pronounced as the sugar content increases. When fine powders are exposed to moisture, the stickiness increases, leading to agglomeration and an overall

deterioration in quality [13]. It should be noted that stickiness, measured with instruments such as texture analyzers (which were also used in this study), provides results that could correlate with sensory evaluation. If the absolute values for stickiness increase, the product can generally be considered stickier [23]. Moisture content is usually considered the main factor affecting the various physical properties of encapsulates such as stickiness, flowability, and generally the quality of the encapsulates and the stability of the active ingredient. Since the encapsulation of plant oils is mainly based on carbohydrates and various biopolymers as carrier systems [22], these compounds are expected to absorb the moisture and contribute to the stickiness after drying [26]. However, the encapsulates produced in this study using the SND encapsulation process have a low moisture content (Table 3), suggesting that moisture content is not the main factor affecting the stickiness of the encapsulates. The main influence on stickiness therefore comes from the oil fraction of the encapsulates (i.e., the oil released during analysis, Table 4).

5. Conclusions

The results of the current work show that the SND encapsulation process developed here can be used as a promising approach for the production of encapsulates loaded with plant oils. Cold-pressed grape seed oil, a known nutrient source, was successfully encapsulated and the encapsulates showed high oil loading. Alginate was used as a carrier material, which ensures good emulsion stability and stable encapsulates. The encapsulates were freeze dried to remove water and increase the shelf life of the encapsulated oil. The obtained dried forms of encapsulates showed similar color characteristics and overall appearance. SEM analysis showed that the encapsulates had an elongated and irregular shape. The hardness and stickiness of the encapsulates were influenced by the oil content. The values of these textural properties indicate that the dried encapsulates are suitable as a carrier for the nutritionally valuable components of cold-pressed grape seed oil to be mixed into a complex food matrix.

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