CHAPTER 8

Microencapsulation of Oils using Whey Protein / Gum Arabic Coacervates*

ABSTRACT
Microencapsulating sunflower oil, lemon and orange oil flavor was investigated using complex coacervation of whey protein / gum arabic. At pH 3.0 – 4.5, the WP/GA complexes formed electrostatic complexes that could be successfully used for microencapsulation purposes. The formation of a smooth biopolymer shell around the oil droplets was achieved at a specific pH (close to 4.0) and the payload of oil (i.e. amount of oil in the capsule) was higher than 80%. Small droplets were easier to encapsulate within a coacervate matrix than large ones, which were present in a typical shell / core structure. The stability of the emulsion made of oil droplets covered with coacervates was strongly pH-dependent. At pH 4.0, the creaming rate of the emulsion was much higher than at other pH values. This phenomenon was investigated by carrying out zeta potential measurements on the mixtures. It seemed that at this specific pH, the zeta potential was close to zero, highlighting the presence of neutral coacervate at the oil / water interface. The influence of pH on the capsule formation was in accordance with previous results on coacervation of whey proteins and gum arabic, i.e. WP/GA coacervates were formed in the same pH window with and without oil, and the pH where the encapsulation seemed to be optimum corresponded to the pH at which the coacervate was the most viscous. Finally, to illustrate the applicability of these new coacervates, the release of flavored capsules incorporated within Gouda cheese showed that large capsules gave stronger release, and the covalently cross-linked capsules showed the lowest release, probably because of a tough dense biopolymer wall which was difficult to break by chewing.

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INTRODUCTION

Microencapsulation techniques are used in food and pharmaceuticals for the protection of enzymes or health ingredients, the taste masking of encapsulated drugs, controlled release, the encapsulation of flavors for food and drinks, and in home and personal care products [see reviews on the subject by: Luzzi, 1970; Dziezak, 1988; Shahidi and Han, 1993; Magdassi and Vinetsky, 1997; Gibbs et al., 1999; Sparks, 1999]. One of the microencapsulation methods is coacervation – phase separation technology, which has been used as a physicochemical procedure for the preparation of polymeric capsules [Deasy, 1984; Arshady, 1990; Ijichi, 1997]. Coacervation was first investigated by Bungenberg de Jong and Kruyt (1929) and was classified into two systems: simple and complex coacervation. The term coacervation is derived from the Latin acervus, meaning “heap” and the prefix co (=together) to indicate gathering of the colloidal particles. Complex coacervation is based on the formation of a complex (coacervate) between oppositely charged polymers, usually proteins and polysaccharides. If the required conditions are met, the polysaccharide and the protein form a complex which may form a film / coating around the target material that needs to be encapsulated [Nairn, 1995]. Traditionally, gelatin is used in combination with gum arabic in numerous studies. The first commercial application was in 1954, when Green and associates of the National Cash Register Company researched the coacervation process for producing carbonless copy paper [Green, 1956]. Since then, this system was applied in many investigations [see for example: Luzzi and Gerraughty, 1964, 1967; Madan et al., 1972, 1974; Newton et al., 1977; Nixon and Nouh, 1978; Takenaka et al., 1980; Takeda et al., 1981; Flores et al., 1992; Jizomoto et al., 1993; Palmieri et al., 1996,1999; Ijichi, 1997; Lamprecht et al., 2000a, 2000b, 2001]. However, nowadays, there is a need to replace gelatin for health and religious reasons. Replacing gelatin by whey protein (WP) and using it in combination with gum arabic (GA) would lead to various advantages. For example, the encapsulation processes with WP/GA coacervation could be carried out at room temperature (or low temperatures > 0°C), which is not possible to achieve with gelatin \( T_{gelation} \approx 40^\circ C \). The encapsulation of temperature - sensitive compounds (e.g. volatile flavors) should be improved with the WP/GA system. Basic studies were already performed on the complex coacervation of WP and GA [Schmitt et al., 1999; Weinbreck et al., 2003a, 2003b] and the conditions under which coacervates are formed are now well known and understood. Renard et al.
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(2002) mentioned the WP/GA system for application in controlled release and encapsulation of drugs, but so far no experimental results on the use of WP/GA coacervates are available. The goal of this work was thus to show that the WP/GA coacervates could be used for encapsulation of oil droplets and flavors. Various conditions influencing the formation of coacervates were studied (pH and biopolymer concentration) and compared to previous results. The stability of the encapsulated emulsion was then investigated as a function of pH. Finally, merely to illustrate the possibilities of the WP/GA coacervates, capsules of lemon oil were incorporated into Gouda cheese and the flavor release was measured in time.

EXPERIMENTAL SECTION

Materials

Bipro is a whey protein isolate (WP) comprised mainly of â-lactoglobulin (â-lg), and á-lactalbumin (á–la) - from Davisco Foods International (Le Sueur, USA). Residual whey protein aggregates were removed by acidification (at pH = 4.75) and centrifugation (1 h at 33000 rpm with a Beckman L8-70M ultracentrifuge, Beckman instruments, The Netherlands). The supernatant was then freeze-dried (in a Modulo 4K freeze-dryer from Edwards High Vacuum International, UK). Finally, the resulting powder was stored at 5°C and it contained (w/w) 88.1% protein (N x 6.38), 9.89% moisture, 0.3% fat and 1.84% ash (0.66% Na⁺, 0.075% K⁺, 0.0086% Mg²⁺ and 0.094%Ca²⁺). The protein content of the treated Bipro is: 14.9% â-la, 1.5% bovine serum albumin (BSA), 74.9% β-lg, and 3.2% immunoglobulin (IMG).

Gum arabic (GA; IRX 40693) was a gift from Colloides Naturels International (Rouen, France). The powder contained (w/w) 90.17% dry solid, 3.44% moisture, 0.338% nitrogen and 3.39% ash (0.044% Na⁺, 0.76% K⁺, 0.20 Mg²⁺ and 0.666% Ca²⁺). Its weight average molar mass (M₆ = 520 000g/mol) and its average radius of gyration (R₉ = 24.4 nm) were determined by size exclusion chromatography followed by multiangle laser light scattering (SEC MALLS). SEC MALLS was carried out using a TSK-Gel 6000 PW + 5000 PW column (Tosoh Corporation, Tokyo, Japan) in combination with a precolumn Guard PW 11. The separation was carried out at 30°C with 0.1 M NaNO₃ as eluent at a flow rate of 1.0 mL min⁻¹.

Stock solutions were prepared by dissolving the powder in deionized water. The pH was adjusted with HCl and NaOH. Three types of oil were used: orange oil flavor from
RC Treatt & Co (Suffolk, England), sunflower oil (Reddy oil) and lemon juice flavor from Givaudan (Dubendorf, Switzerland). Glutaraldehyde grade II 25% was purchased from Sigma.

**Preparation of the capsules**

The capsules were always prepared in the same way at room temperature, independently of the type of oil used. First, an oil in water (O/W) emulsion was prepared by dispersing the oil into a stock solution of WP at pH 7. Then, a stock solution of GA (at pH 7) was poured into the O/W emulsion and stirred for 30 min. The final concentration of oil was 5% (w/w) in all experiments (after the addition of the GA solution). The ratio of WP to GA (Pr:Ps) was kept constant at 2:1 (w/w), since most of the previous work was done at this ratio [Weinbreck *et al.*, 2003a]. The pH of the mixtures was adjusted to the desired value and the mixture was left under slow stirring overnight.

**Influence of physico-chemical parameters (pH, biopolymer concentration, droplet size)**

Encapsulated orange oil was prepared as described above. The influence of various parameters on the encapsulation of oil was investigated: pH, total biopolymer concentration (Cp) and droplet size. Various pH values were studied (from 2.0 to 5.0 in 0.5 steps and at pH 7.0) for Cp = 1%. Cp was varied from 0.05 % to 3.0 % (w/w) at pH = 4.0. The oil droplet size was varied by dispersing the oil with a blender (Polytron, Kinematica, Switzerland) at speed 4 for 5 min, or with a magnetic stirrer (Cp = 1% and pH = 4.0). The size of the oil droplets was measured with a Mastersizer X (Malvern, UK). The size of large droplets (prepared with a magnetic stirrer) was very polydisperse and varied from 50 μm to 1000 μm. Emulsions prepared with the blender had a size between 5 μm and 50 μm. The capsules were visualized under a light microscope (Leica, Rijswijk, The Netherlands).

**Emulsion stability and zeta potential**

Encapsulated sunflower oil was prepared as described above (Cp = 1%), except that after emulsifying the oil with a blender, the emulsion was homogenized at 200 bar with a Rannie homogenizer (type 8.30 H) to obtain a stable emulsion at pH 7. The droplet size measured with the Mastersizer X was around 1.5 μm. The pH of the emulsion was
varied from pH 2.0 to 7.0. The turbidity of the emulsion was measured several times per day with a Turbiscan (Formulaction, France) during 5 days. This instrument measures the intensity of the back-scattered light along the height of the 1 cm diameter glass tubes containing the emulsions. The creaming rate of the emulsion could thus be determined by following in time the leveling off of the interface between the lower clear layer and the upper cream layer.

The zeta potential of the homogenized emulsion of encapsulated sunflower oil (Cp = 1%, 5% sunflower oil, and pressure = 200 bar) was measured as a function of pH. The titration was carried out with 1 M HNO$_3$ from pH 7 to pH 2. Two blanks consisting of an emulsion homogenized only with WP and an emulsion homogenized only with GA were prepared and their zeta potential was also measured as a function of pH. The measurements were made with an Acoustisizer (Matec Applied Sciences Inc., Hokinton, USA) at Philips Natlab in Eindhoven, The Netherlands.

**Encapsulated lemon oil in Gouda cheese**

Lemon flavor was encapsulated following the same procedure as used for encapsulated sunflower oil (described above), but the process was carried out at 5°C to limit flavor evaporation. The amount of oil was set at 6.67%, Cp = 1%, pH = 4.0. One batch was made by dispersing the oil with a blender (small droplets) and another one with a magnetic stirrer (large droplets). From each batch, blanks of the emulsion were prepared at pH 7.0. At that pH, no coacervation occurred and the oil was not encapsulated. From the encapsulated batch (at pH 4.0), one part was not cross-linked and the other part was cross-linked by slowly adding concentrated glutaraldehyde to the mixture with stirring, in order to reach a final glutaraldehyde concentration of 0.1%. The mixtures were then mixed for 45 min; the capsules were washed to get rid off the residual glutaraldehyde and filtered. The slurry was redispersed in deionized water to reach the initial volume of the mixture. The capsules were added to Gouda cheese during the cheese process at NIZO food research. 5 ml of each mixture (blank, non-cross-linked capsules and cross-linked capsules – small and large capsules) were added to 1800 g of curd and the curd was mixed by hand for 1 min. Afterwards, the curd was pressed into four identical cheeses of 250 g each, brined (4 h), and graded for one month. After one month, the cheeses were tasted in duplicate by five panelists with the MS Nose. The panelists were instructed to chew the cheese regularly for 30 s without swallowing, then to swallow the entire piece of cheese, and after swallowing, to
continue chewing for 60 s. The order of the samples was randomized. While panelists were eating the cheese, the nose space concentration of limonene (main component of lemon oil) was monitored by sampling the airflow from one nostril over a 1.5 min period. A full description of the apparatus is available in Weel et al. (2002). An average of the limonene intensity from all panelists’ results was calculated for the six cheeses and plotted as a function of time.

RESULTS AND DISCUSSION

Influence of physico-chemical parameters (pH, biopolymer concentration, droplet size)

Microencapsulation of oil droplets by the complex coacervation method is known to be strongly influenced by many parameters, such as pH, biopolymer concentration (Cp), and droplet size [Madan et al., 1972, 1974; Nixon and Nouh, 1978; Takenaka et al., 1980; Burgess and Carless, 1985; Arshady, 1990; Burgess, 1994; Ijichi et al., 1997; Thimma and Tammishetti, 2003]. Since the formation of complex coacervates arises from electrostatic interactions between the positively charged WP and the negatively charged GA, coacervates can be formed in the pH range 2.5 – 4.8 [Weinbreck et al., 2003a]. Orange oil droplets were encapsulated with WP/GA coacervates following the method described above. The microcapsules were made at various pH values and were observed under the light microscope to see if a coacervate wall was visible around the oil droplets (Figure 8.1). It appeared that at pH > 5.0, no encapsulation took place. In the pH region 4.5 – 3.0, a layer of coacervate was noticeable around the oil droplets and the capsules formed had the tendency to stick together in the absence of shear. At pH 4.5, coacervates were visible in the mixture encapsulating partially some oil droplets and partially in the bulk solution. At pH 4.0, a smooth coacervate layer homogeneously enveloped the oil and the capsules formed clusters when stirring was stopped. At pH 3.5, the oil was still well encapsulated but the coacervate layer seemed less homogeneous. At lower pH (below 3.0), the oil droplets were not encapsulated. These results were in good agreement with previous studies [Weinbreck et al., 2003a]. From that work, it was found that complex coacervation between WP and GA occurred in a specific pH window, which was the same when oil was added to the mixture. The viscosity of the coacervate layer was previously investigated and was found to be very much pH-dependent [Weinbreck et al., 2004b]. Here, the best capsules were formed
at pH 4.0, at which the viscosity of the coacervate was the highest and at which most polymers were present in a coacervate form [Weinbreck et al., 2004b]. Burgess (1994) reported previously that a high viscosity of the coacervate resulted in more stable capsules, but if the viscosity was too high, then encapsulation could be impaired. Each biopolymer combination needs to be investigated to obtain the optimum conditions for encapsulation. For WP/GA, pH = 4.0 seemed to give the best results at a Pr:Ps of 2:1.
If the ratio of protein to polysaccharide is changed, then the optimum pH would also be shifted [Weinbreck et al., 2003b].

The influence of total biopolymer concentration was studied on a 5% oil in water (O/W) emulsion. The Cp was varied from 0.1% to 3.0% and the pH was set at 4.0. The samples were visualized under the microscope (Figure 8.2). A thin and homogeneous coacervate layer was observed around the oil droplets at Cp > 0.5%. For Cp > 1%, the coacervate layer was thicker and free coacervates were found in solution. A Cp between 0.5% and 1% seemed to be sufficient to encapsulate 5% of oil. The payload of oil: polymer would be between 90% and 80%. This high payload is a major advantage of encapsulation using complex coacervation and is in the same range if gelatin is used [Luzzi, 1970; Dziezak, 1988]. For Cp > 8%, one would expect a reduction of the amount of coacervate because of a self-suppression on coacervation at high concentrations [Burgess and Carless, 1985; Burgess, 1994; Weinbreck et al., 2003a].

Various oil droplet sizes could be achieved, depending on the way the emulsion was prepared. Small droplets (D_{3,2} = 5 – 50 μm) were made by using a blender and large droplets (D_{3,2} = 50 – 200 μm) by dispersing the oil with a magnetic stirrer. In both cases, the emulsions were very polydisperse. From microscopic observations, it was found that small droplets were easily encapsulated whereas some large droplets were only partially or not at all encapsulated. The capsules also looked different depending on the size of the oil droplets. Large droplets were individually surrounded by a thin coacervate layer, whereas small droplets were encapsulated in a matrix of coacervate as shown in Figure 8.3. It has already been reported that oil droplets larger than 250μm

Figure 8.3: Encapsulated orange flavor with WP / GA coacervates, 5% oil, Cp = 1%, pH = 4.0, Pr:Ps = 2:1. (a): large droplets prepared with magnetic stirrer; (b): small droplets prepared with a blender. The scale bar represents 200 μm.
were difficult to encapsulate [Madan et al., 1972; Ijichi et al., 1997]. This difference might be attributed to a different encapsulation mechanism for small and large droplets. Upon pH decrease, coacervate droplets are initially formed in the bulk, and then they migrate, wet the surface of the oil and spread [Madan et al., 1972; Ijichi et al., 1997]. The wetting and spreading of the coacervate has a major impact on the encapsulation [Madan et al., 1972; Newton et al., 1977; Ijichi et al., 1997; Thomassin et al., 1997]. Good wetting properties of the coacervate are required and the viscosity of the coacervate should be high enough to form a resistant barrier around the oil droplets but not so high as to prevent spreading on the surface. The encapsulation of large droplets requires the aggregation and the partial coalescence of a great amount of coacervate droplets. On the other hand, for encapsulation of small droplets, entrapment of the substance in one coacervate droplet is more likely to occur and requires less rearrangement than wetting and spreading, which involves a change of shape of the coacervate.

**Stability of the encapsulated emulsion**

When oil droplets were encapsulated by complex coacervation of WP and GA, it was noticed that the emulsion was highly unstable against creaming. The stability of a homogenized emulsion ($D_{[3,2]} = 1.5 \text{ m}$) for $C_p = 1\%$ was investigated as a function of pH. Creaming of the oil droplets was measured with the Turbiscan. The interface between the clear lower phase and the cream layer was sharp and evolved in time. A sample at pH 7 was taken as a reference. Only after 5 days a small cream layer was measurable. From these measurements, the volume of the clear layer could be plotted as a function of time, and the initial slope gave the velocity of creaming ($s$). The creaming velocity at pH 7.0 ($s_0$) was very low, $s_0 = 0.0002 \text{ mm.min}^{-1}$. In Figure 8.4, the relative velocities ($s/s_0$) of all samples are plotted as a function of pH. The instability of the emulsion was strongly pH-dependent. For pH 6.0, no creaming was observed after 2 days ($s/s_0 = 1$). By decreasing the pH from 6.0 to 3.6, the creaming rate increased and a maximum was measured at pH 3.6, where the creaming was 400 times faster than at pH 7. Below pH 3.6, $s/s_0$ decreased. Since the electrostatic interaction between WP and GA is strongly pH-dependent, the formation of a WP/GA complex at the oil / water interface decreased and the repulsion between droplets and creaming was enhanced. Zeta potentials of the emulsion stabilized with WP and GA were measured as a function of pH in order to quantify the charge profile of the oil.
droplets. Blanks of emulsions stabilized only with WP or only with GA were also carried out. The results are shown in Figure 8.5. The emulsion stabilized with only GA had a negative Helmhotlz - Smoluchowski potential from pH 7.0 until pH 2.5 and the emulsion stabilized with only WP had a negative potential above its pI (= 5.2) and a positive potential below. The results of the blanks were as expected. Indeed, the pI of â-lg (the main protein present in WP) is known to be around this value [Weinbreck et al., 2003a]. GA is a weak polyelectrolyte and its charge density decreases at low pH, as was already measured in a bulk solution [Burgess and Carless, 1984; Schmitt et al., 1999] or as an emulsifier [Jayme et al., 1999]. The Helmhotlz - Smoluchowski potential was then measured for an emulsion stabilized with WP and GA as a function of pH. For pH > 5.3, the zeta potential of the emulsion was negative, as for an emulsion emulsified only with WP. At pH 5.0, the potential of the emulsion decreased to the same value as the potential of the emulsion made only with GA. From pH 5.0 to pH 3.0, the potential of the mixed emulsion was very close to zero. At pH 2.7 and below, the potential of the mixed emulsion increased drastically to reach high positive values. The results of the zeta potential measurements were in good agreement with the values of the emulsion stability and with the previous results on the behavior of WP/GA as a function of pH.
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Figure 8.5: Helmholtz - Smoluchowski potential of a 5% sunflower oil emulsion as a function of pH; ( ): emulsion stabilized with WP; (∇): emulsion stabilized with GA; ( ): emulsion stabilized with WP/GA.

[Weinbreck et al., 2003a]. Since WP was negatively charged above pH 5.2, GA (also negatively charged) did not interact with the WP, repulsion occurred between the oil droplets and the emulsion remained stable. Just below pH 5.2, the WP became positive and interacted with the GA present in the mixture. It has already been shown in previous studies that soluble complexes of WP / GA were formed in the pH region 5.2 – 4.8 [Weinbreck et al., 2003a]. As a result, the zeta potential of the emulsion was similar to the zeta potential of the blank emulsion homogenized with GA. Between pH 4.8 and 2.5, coacervates were formed as shown by Weinbreck et al. (2003a) and a coacervate layer was formed around the oil droplets (c.f. Figure 8.1). The coacervate layer being a quasi-neutral phase, the zeta potential measured was close to zero, and a strong attraction between oil droplets occurred, which was responsible for a fast creaming rate of the emulsion (cf Figure 8.4). Then, below pH 2.5, GA, being a weak polyelectrolyte, lost its charge and reached neutrality. As a consequence, the coacervates were disrupted and the oil / water interface was probably composed of some neutral GA and certainly composed of positive WP, as indicated by the positive zeta potential of the emulsion and the decrease of creaming rate. For preparing good encapsulated material, it is thus very important to maintain stirring during the whole process to avoid
phase separation. To avoid the clotting and creaming of the encapsulated droplets, the coacervate layer can be cross-linked chemically (e.g. glutaraldehyde) or enzymatically (e.g. transglutaminase). The coacervation then becomes irreversible, WP and GA being covalently cross-linked, the pH of the mixture can then be varied (above pH 6.0) and repulsion between droplets occurs without breaking the coacervate layer.

**Encapsulated lemon oil in cheese**

Large capsules (size ~ 50 – 1000 μm) and small capsules (size ~ 5 – 50 μm) of lemon oil were prepared by complex coacervation of WP/GA. One batch was cross-linked with 0.1% glutaraldehyde and the other batch was not cross-linked. Blanks were made of the same emulsion of lemon oil with WP and GA but at pH 7 (where no capsules were formed) and they were compared to the capsules. The capsules were put in a Gouda cheese, by incorporating them into the curd during the cheese process. The cheeses were graded for one month and the release of the flavor was monitored with the MS Nose. The six cheese samples containing the capsules were tasted by five panelists and the amount of limonene released was measured in real time. The average flavor

![Figure 8.6: Average flavor release, MS Nose curves from cheeses containing limonene capsules. (A): Large capsule, blank pH 7; (B): Large capsule, encapsulated in WP/GA pH 4, not cross-linked; (C): Small capsule, encapsulated in WP/GA pH 4, not cross-linked; (D): Small capsule, blank pH 7; (E): Small capsule, encapsulated in WP/GA pH 4, cross-linked; (F): Large capsule, encapsulated in WP/GA pH 4, cross-linked.](image-url)
release was plotted in Figure 8.6. All panelists’ results were similar. The larger capsules (blank and non-cross-linked) gave the strongest flavor release. The cross-linked batches (small and large capsules) gave the lowest release intensity. The time at which the maximum intensity was detected is plotted in Figure 8.7. The maximum intensity was reached at approximately the same time for all samples, just after swallowing took place ($t_{\text{swallowing}} = 0.5$ min). The largest capsules gave the strongest release, probably because it was easier the break them upon chewing compared to the small capsules. The fact that the cross-linked capsules gave a low release could be explained if not all capsules were broken upon chewing, the cross-linking giving very strong capsule walls. However, it should be mentioned that the preparation of cross-linked capsules included an extra washing step, in which part of the flavor could have been washed away, but it is believed that this effect is very small. Preliminary studies on the degree of flavor evaporation by gas chromatography showed that the evaporation was delayed by a factor of 4 for non-cross-linked (large) capsules and by a factor of 10 for cross-linked capsules. The amount of limonene in the capsules and its oxidation products should be studied in more detail in further shelf-life experiments in order to get a more quantitative
description. So far, it has been shown that encapsulation with WP/GA is feasible. To improve the release of flavor in a product, a large droplet size is preferred and the degree of cross-linking should be carefully balanced between a well-encapsulated material and a high release.

CONCLUSIONS

Capsules containing oil (e.g. flavors) could successfully be prepared using complex coacervation of WP and GA under specific conditions. Parameters such as pH, polymer concentration and capsule size were investigated and the results were in very good agreement with previous basic work. Capsules prepared at pH 4.0 showed a smooth shell of WP/GA coacervate around the oil droplets. pH 4.0 corresponds to the pH at which all WP and GA reacted to form a coacervate. Furthermore, at this pH, the strength of the electrostatic interaction and the viscosity of the coacervates were at maximum. A high payload (up to 90%) of oil versus polymer could be achieved. Small oil droplets (< 50 μm) could be encapsulated into a coacervate matrix more easily than large droplets. Large oil droplets (> 200 μm) were formed by a typical core / shell mechanism and were occasionally only partially encapsulated. For preparation of the capsules at pH between 4.5 and 3.0, it was very important to use continuous stirring. Indeed, emulsions stabilized with WP/GA coacervates were highly unstable against creaming at pH < 5.0. The fastest creaming rate was obtained at pH 3.6 and corresponded to a surface charge that was close to zero. As an example, various capsules of lemon oil were prepared and introduced into a cheese. After one month, the flavor release was higher for large capsules, which were easier to chew, than small ones. The degree of cross-linking should be carefully tuned since cross-linked capsules gave a lower flavor release than the non-cross-linked batch. These results showed that a matrix of WP / GA can be successfully used for encapsulation purposes. However, more work is needed to check the barrier properties of these coacervates against water, flavor, and oxygen diffusion.

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