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Original article

Effect of pectin and temperature on sunflower meal colloidal particles for emulsion structure and stabilisation

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Summary This study evaluated the formation and dispersion effects with 4% w/w total biopolymer [sunflower meal protein (SMP) and pectin (P)] at SMP:P ratio 1:0, 25:1, 15:1, 5:1, pH 3.7, prepared at room temperature (RT) or heat treated (HT) at 98 °C on emulsion stabilisation. At RT, increasing P concentration increased apparent viscosity and gel-like behaviour of biopolymer dispersions and emulsions. At SMP:P-RT ratio 5:1, the dispersion structure showed protein attached to pectin and individual pectin dispersed in the continuous phase. Under this condition, the emulsion presented stability regarding creaming, flocculation and/or coalescence. Whereas when HT, all samples showed low apparent viscosity, but at SMP:P ratio 25:1 and 15:1 emulsions exhibit gel-like behaviour. However, SMP:P ratio 5:1 dispersion presented spheroid particles and emulsion behaved as liquid-like material with lower creaming, flocculation and/or coalescence. Therefore, depending on the process parameters, sunflower meal may be used as an ingredient to obtain different food emulsion structure.

Keywords Electrostatic complexes, emulsion stability, polysaccharide, rheological properties, sunflower.

Introduction

The demand and interest in plant-based proteins have been extensively highlighted due to the growing worldwide population, lower environmental impacts on its obtention and lower production costs (Sá *et al.,* 2020). The use of industrial side streams and by-products to produce food products presents an efficient way to lower both costs and environmental impacts of the production (Heusala *et al.,* 2020). However, it is necessary to develop methods for plant-based food ingredients production with improved sensory, nutritional and functional properties through research innovation (Day, 2013).

Sunflower seed production has been growing especially for oil extraction, and its by-product, sunflower meal (SM), has a high protein concentration (25–50 wt%), depending on the oil extraction process and sunflower seed variety (González-Pérez & Vereijken, 2007; Pickardt *et al.,* 2015). SM is mainly intended for animal feed due to the loss of protein functional properties in the oil extraction process and the presence of chlorogenic acid (Kreps *et al.,* 2014).

The functional properties of proteins contribute in many ways to the technological characteristics of products, such as texture and stability of foams and emulsions. Emulsifying properties are related to the surface activity of proteins at oil–water interface, reducing interfacial tension and forming cohesive films around the oil droplets. The instability mechanisms to be controlled in emulsions are creaming, flocculation and coalescence. Creaming occurs due to the density difference leading to oil droplets concentrating on the surface and phase separation. Flocculation is related to the forces of attraction between the droplets, causing their physical aggregation, and coalescence corresponds to the union of two or more nearby oil droplets, forming one larger droplet (Walstra & Vliet, 2008; Day, 2013).

Besides proteins, polysaccharides are widely used in food, especially for their gelling and stabilising properties. Pectin is an anionic polysaccharide used for its effects on

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the texture and stabilisation of products (Yuliarti *et al.,* 2019). It is a high molecular weight linear chain of α-1,4-d-galacturonic acid units and its gelling properties depend on factors such as pH, temperature and degree of esterification (DE) (Chan *et al.,* 2017).

Mixtures of proteins and polysaccharides may produce improvements in functional properties compared to their individual use (Dickinson, 2008), and the characteristics of the particles formed from these mixtures depend on the conditions in which they are produced. Electrostatic complexes could be formed between proteins and polysaccharides with opposite charge. Therefore, the protein pH should be adjusted to a value near or below the isoelectric point (pI) if an anionic polysaccharide is used or pH ≥ pI of proteins, if a cationic polysaccharide is used (Patino & Pilosof, 2011).

Studies have been carried out to evaluate the influence of different composition and process conditions, such as pH, protein:polysaccharide ratio and heat treatment, on the formation and characteristics of electrostatic protein–polysaccharide complexes (Stounbjerg *et al.,* 2019; Lan *et al.,* 2020; Wang *et al.,* 2022). These electrostatic complexes can be used to improve emulsifying properties (Wang *et al.,* 2019; Kotchabhakdi & Vardhanabhuti, 2020; Zhang *et al.,* 2022).

Therefore, the aim of this study was to evaluate the influence of the pectin addition and the heat treatment on the formation and structure of electrostatic complexes between SM protein and pectin and their effect on emulsion rheology and stability.

Materials and methods

Materials

Sunflower meal, obtained from a commercial mixture of seeds by grinding the cake after oil extraction, was patented in 2019 (EP2400859) and donated by the Fraunhofer Institute, IVV (Freising, Germany) as part of the bilateral Brazil–Germany cooperation project 'Sunflower Protein' (Project number: 031A281A). The meal composition was 6.51 ± 0.13% of moisture, 8.32 ± 0.02% of ash and 53.01 ± 0.13% of protein (N × 6.25), by the AOAC (1997) methodologies; fat content was 1.50 ± 0.12% by the method of Bligh & Dyer (1959); and carbohydrates were 30.66%, obtained by difference. Thermograms, obtained from 20 to 120 °C at a heating rate of 1.0 °C min^{-1} in a MicroCal VP-DSC (Malvern Instruments, Worcestershire, UK), showed two transitions at temperatures at approximately 75 and 90 °C.

Low-methoxy pectin (GENU Pectin type LM CG-22, DE 47.2%, molecular mass 90 kDa) was donated by CPKelco (Grossenbrode, Germany). Reagents were of analytical grade and sunflower oil was purchased in local market.

Preparation and characterisation of biopolymer dispersions

Sunflower meal and pectin (P) samples were dispersed in deionised water by stirring at room temperature (RT) and stored under refrigeration overnight, for complete hydration. Then SM and P dispersions were mixed in a 1:0, 25:1, 15:1 and 5:1 SMP:P ratios (w/w), where SMP corresponds to SM proteins. All of the mixtures were prepared with 4% w/w total biopolymer concentration, where biopolymer corresponded to proteins, pectin or their mixture. The pH was adjusted to 3.7 with 1 M HCl and the ionic strength 0.06 with NaCl. The samples were prepared at RT and heat treated (HT). For samples HT, the dispersions were heated in an oil bath until temperature reached 98 °C (10–15 min) and immediately cooled to RT.

Zeta potential

Zeta potential of SM (4% w/w) and P (0.08% w/w) and their mixtures was measured using the Zetasizer Nano Series equipment (Malvern Instruments). Dispersions were prepared at RT with Milli-Q water, filtered through ordinary filter paper and the pH values were adjusted in the range 2.0–8.0 with 0.1 or 1 M HCl or NaOH.

Microstructure

Atomic force microscopy (AFM) WITec (alpha 500, Ulm, Germany) was used to display the topography of the biopolymer dispersions. The samples (2–3 μL of 5 μg mL^{-1} dispersion) were deposited on glass slides and then dried at RT for 120 min.

Emulsion preparation and characterisation

Emulsions were prepared at 1:3 ratio (w/w) (sunflower oil:biopolymer dispersions at different ratios), as previously indicated (item 2.2) and homogenised at RT using a Ultraturrax equipment (IKA-Werke GmbH & Co, Staufen, Germany) at 15 500 rpm for 5 min.

Droplet size distribution

The droplet size distribution of the emulsions was evaluated using a laser diffraction particle size analyser and deionised water as a dispersant (Horiba LA-950, Horiba Ltd, Inc., Kyoto, Japan). The droplet size was expressed as mode, which is the peak of the frequency distribution.

Emulsions stability

Emulsion stability was monitored by vertical scan analyser Turbiscan MA 2000 (Formulaction, Toulouse, France). Samples of freshly prepared emulsions were placed into the glass cylindrical tube and scanned from bottom to top. Transmission (%T) and backscattering

(%BS) measurements were recorded every minute during the first 3 h and every 24 h for the next 4 days (Camino & Pilosof, 2011). Samples were stored at 20 °C.

Rheology of biopolymer dispersions and emulsions

Viscosity and viscoelastic properties of the biopolymer dispersions and emulsions were evaluated using a controlled stress rheometer (Anton Paar GmbH, Graz, Austria) equipped with a coaxial cylinder (inner diameter 26.7 mm, outer diameter 28.9 mm). Apparent viscosity was measured by varying the shear rate of 0–300 s^{-1} and it was evaluated at 15.8 s^{-1}. The results were fitted to power law equation, $\sigma = k\gamma^n$, where σ is shear stress, k is the consistency index, γ is the shear rate and n is the flow behaviour index.

The storage modulus (G') and the loss modulus (G'') were recorded while the frequency sweep ranged from 0.1 to 100 rad s^{-1} with strain of 1% for the SM:P ratio 5:1 at 98 °C sample and at 0.1% strain for other samples. Analysis were conducted at 20 °C and repeated at least three times for each dispersion and emulsion preparation.

Statistical analysis

The droplet size distribution results were evaluated statistically by analysis of variance and Tukey test ($P < 0.05$).

Results and discussion

Biopolymer dispersions

Figure 1 shows the zeta potential profiles of the dispersions from SM, P and for different SMP:P ratios, as a function of pH.

Sunflower meal presented a net charge close to zero within the pH range between 2.0 and 4.0, reaching point of zero charge around pH 3.0 and negative charge above pH 4.0 (Fig. 1). Subaşı *et al.* (2020) reported for sunflower protein isolates natural (80% protein) and dephenolized (87% protein) pI 4.4 and 4.8, respectively. Other components such as carbohydrates and chlorogenic acid in the SM used in this study may have affected the electric charge balance, leading to pI values lower than those reported in literature. Karefyllakis *et al.* (2019) studied mixtures from sunflower seeds and obtained point of zero charge at pH 3.5–4.0 for protein:fibre ratio 2:5, whereas the sample protein:fibre ratio 2:8 did not reach the *x*-axis, presenting negative charge. González-Pérez & Vereijken (2007) highlighted that composition of sunflower seed varieties and the SM extraction method modify the pI. Pectin showed negative charge along the curve with pKa close to 2.0 (Fig. 1).

According to Fig. 1, the zeta potential profile of the mixture SMP:P ratio 25:1 was closer to SM indicating the predominance of protein charges. Increasing the concentration of P (SMP:P ratio 25:1 to 5:1), most of the positively charged protein interacted with

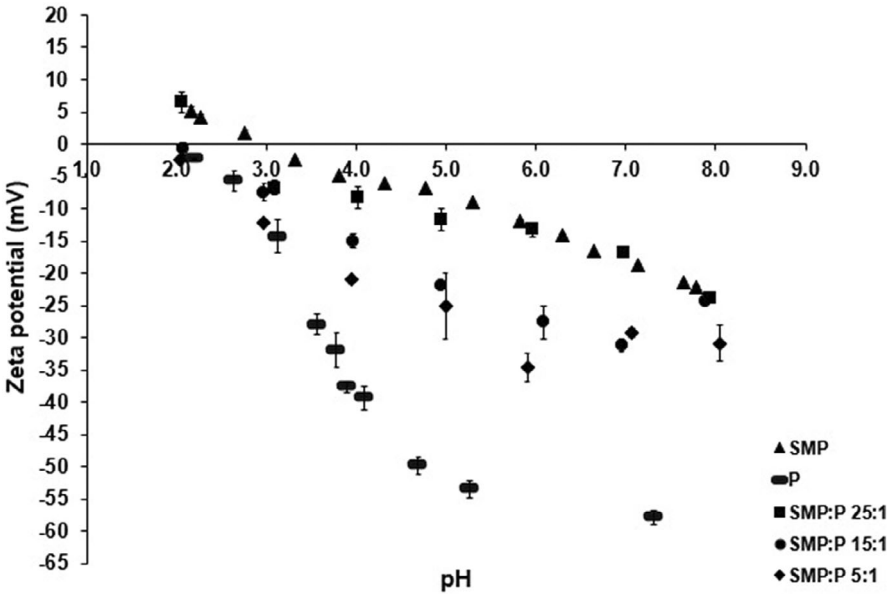


Figure 1 Zeta potential (mV) as a function of pH for dispersions of sunflower meal (SM), pectin (P) and SMP:P ratios of 25:1, 15:1 and 5:1 (w/w), where SMP corresponds to SM proteins.

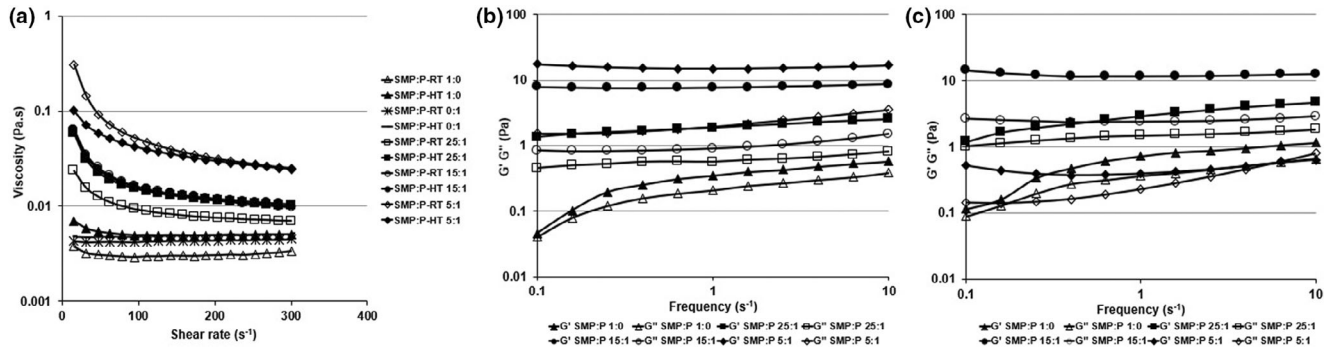


Figure 2 (a) Apparent viscosity as a function of shear rate (s^{-1}) of sunflower meal (SM), pectin (P) and dispersions SMP:P ratio 1:0, 0:1, 25:1, 15:1 and 5:1 (w/w), where SMP corresponds to SM proteins, prepared at room temperature (RT, open symbol) and heat treated (HT, filled symbol); elastic modulus (G', filled symbol) and viscous modulus (G'', open symbol) as a function of frequency (s^{-1}) of dispersions SM:P ratio 1:0, 25:1, 15:1 and 5:1 prepared at (b) RT and (c) HT.

negatively charged pectin and the dispersions showed higher negative charge at the same pH.

Rheological properties are shown in Fig. 2. Viscosity behaviour of the dispersions as a function of shear rate was evaluated (Fig. 2a) and the parameters k (consistency index) and n (flow behaviour index) were obtained from the power law model ($\sigma = k\gamma^n$) (Table S1).

Dispersions of SM and P (SMP:P ratio 1:0 and 0:1, respectively), prepared at RT and HT, presented values of apparent viscosity independent of shear rate, indicating Newtonian behaviour (Fig. 2a). With P addition, the dispersions SMP:P-RT ratio 25:1, 15:1 and 5:1 showed shear thinning behaviour ($n < 1$) (Table S1), or values of apparent viscosity dependent on the shear rate, suggesting the interactions and structure between biopolymers are aligned with the shear field. The greater formation of electrostatic complexes at higher P concentration, indicated by higher electrostatic repulsion according to zeta potential values (Fig. 1) led to an increase in the apparent viscosity of the dispersions (Fig. 2a). Comparing dispersions with the same SMP:P ratio, the HT promoted an increase in apparent viscosity for dispersions ratio 1:0 and 25:1, whereas at ratio 15:1 no change was observed; SMP:P ratio 5:1 showed decrease in viscosity (Fig. 2a). Therefore, the dispersions viscosity decreased with the increase in P concentration when HT.

The viscoelastic properties of SMP:P-RT and SMP:P-HT dispersions are presented in Fig. 2b and c, respectively. All samples prepared at RT showed $G' > G''$ values indicating the prevailing gel structure (Fig. 2b). The dispersion SMP:P-RT ratio 1:0 showed a viscoelastic behaviour, the frequency-dependent values of G' and G''. On the other hand, the dispersions with SMP:P-RT ratio 15:1 and 5:1 also presented $G' > G''$, but G' values were independent of frequency that indicates the formation of an interconnected gel-like network structure and solid material characteristics. Figure 2c shows that the G' and G'' behaviour of dispersions subjected to HT was similar to dispersions prepared at RT (Fig. 2b), although the values were, in general, higher, which suggested the strengthening of the structure. During heating, the electrostatic interaction between pectin and protein is less strong due to weakly charged pectin; hence, proteins could form aggregates by hydrophobic and disulphide bonds onto which pectin was adsorbed by hydrophobic interactions and hydrogen bonds (Jones *et al.,* 2010a; Kayitmazer, 2017; Pathak *et al.,* 2017). Nevertheless, for dispersion SMP:P-HT ratio 5:1, values of G' and G'' were lower than the same dispersion prepared at RT, indicating a viscous behaviour. Therefore, higher protein content led to higher viscoelasticity with stronger gel-like network structure due to the formation of electrostatic interaction between protein and pectin (Lan *et al.,* 2020).

Figure 3 shows the images by AFM of the dispersions SMP:P ratio 1:0, 25:1, 15:1 and 5:1 prepared at RT and HT. From SMP:P-RT ratio 1:0 to 15:1, increasing P concentration led to particle size increase, confirming the electrostatic interaction between SMP and P, as discussed before. These results corroborate the viscosity and viscoelastic properties since both explain the structure of the dispersions. The image of the dispersion SMP:P-RT ratio 5:1 appeared grainy suggesting protein attached to pectin chain by electrostatic interactions with individual P chain in the continuous phase (Fig. 3). Whereas, after HT, the electrostatic complexes showed spheroid particles without individual P chain in the continuous phase. This structure led to viscous behaviour and lower apparent viscosity for dispersion SMP:P-HT ratio 5:1 compared to those prepared at RT. The dispersion SMP:P-HT ratio 5:1 resulted in the formation of some aggregates (Fig. 3), which may be a consequence of the use of temperatures above the temperature of thermal denaturation of the globular protein (Jones *et al.,* 2010b).

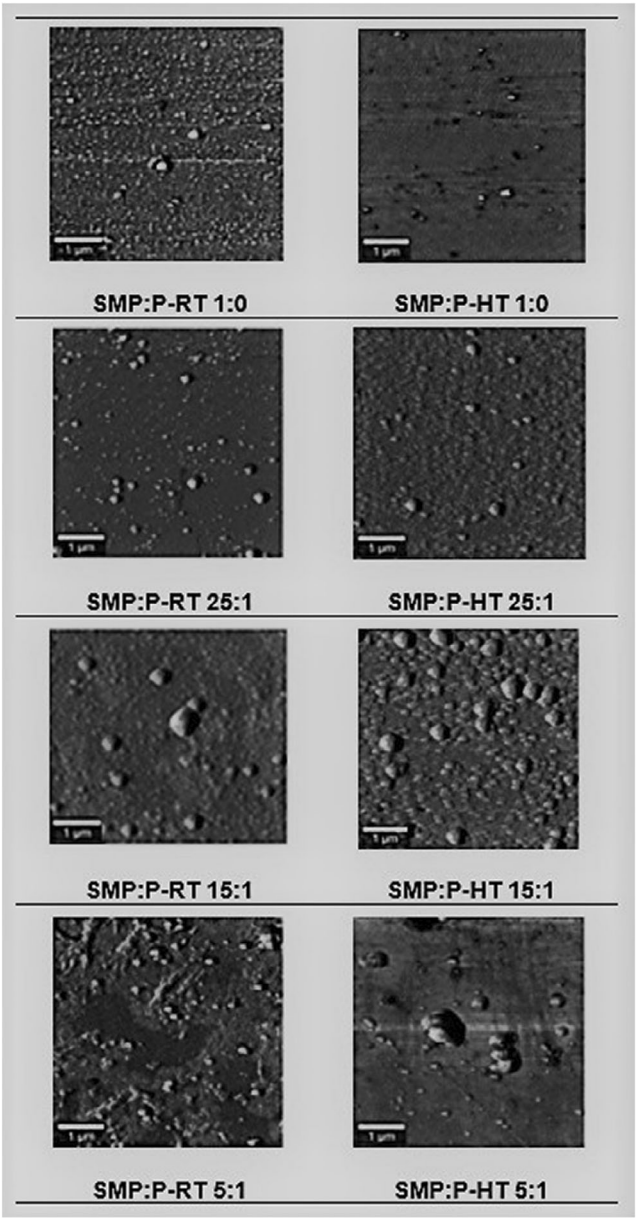


Figure 3 Atomic force microscopy image of sunflower meal (SM) and pectin (P) dispersions, SMP:P ratio 1:0, 25:1, 15:1 and 5:1 (w/w), where SMP corresponds to SM proteins, prepared at room temperature (RT) and heat treated (HT). Scale: 1 μm.

Emulsions characterisation and stability

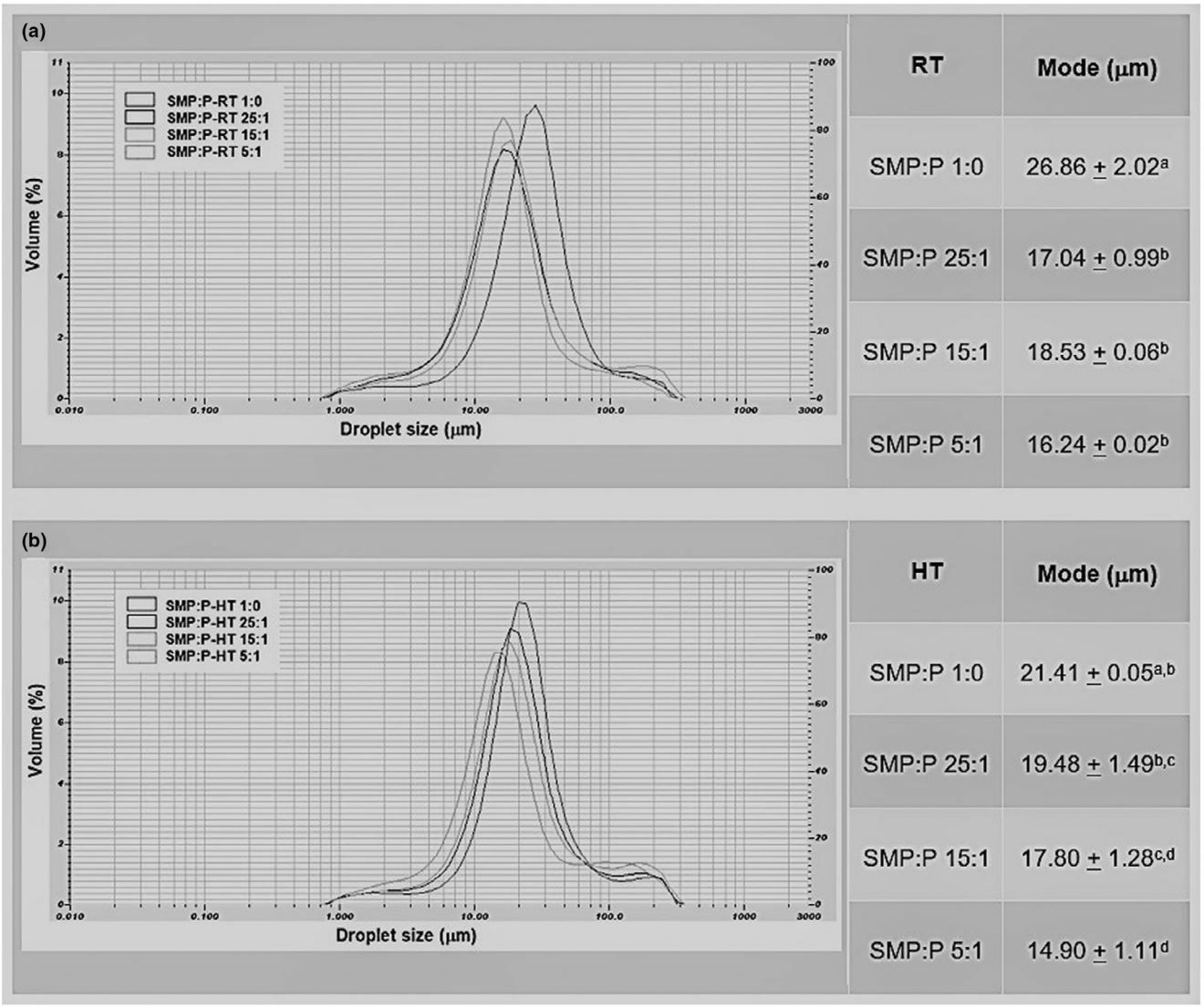
The droplet size distribution of the emulsion prepared with dispersions at different SMP:P ratios showed monomodal distribution, with a wide diameter range and a peak mode between 10 and 20 μm (Fig. 4).

The emulsions with SMP:P-RT ratio 1:0 and SMP:P-HT ratio 1:0 presented the highest frequency of larger droplets diameter. In general, emulsions prepared with dispersions containing P presented a displacement of droplet size distribution curves towards smaller diameters (Fig. 4). Emulsion prepared with HT dispersion presented lower apparent viscosity leading to droplet size reduction (Fig. 5a).

The viscoelastic properties of emulsions were assessed by recording the G' and G'' as a function of the oscillation frequency (ω) within the linear viscoelastic range. Loss tangent, $\tan\delta = (G''/G')$, was calculated from these data. According to Fig. 5b, all samples showed gel-like behaviour ($\tan\delta < 1$ or $\delta < 45°$) with the exception of the emulsion prepared with SMP:P-HT ratio 5:1, which behaved as liquid-like ($\tan\delta > 1$ or $\delta > 45°$) material over the frequency range. This result could be explained by the viscous behaviour and low apparent viscosity of the dispersion SMP-P HT ratio 5:1, which result in an emulsion with a reduced droplet size.

To evaluate the emulsions stability, transmission (%T) and backscattering (%BS) profiles as a function of the height of the cylindrical glass cell (80 mm), from bottom to top, and of the time (during the first 3 h and every 24 h for 4 days) were collected (Figure S1). In the left side of %T profile, values >50%, from emulsions prepared with dispersions SMP:P-RT ratios 1:0 and 25:1 and SMP:P-HT ratios 1:0, 25:1 and 15:1 (Figure S1a–d and f), and close to 0%T, from emulsion prepared with dispersion SMP:P-HT ratio 5:1 (Figure S1h), represented changes at the bottom of the glass cell (0–25 mm) and, therefore, the occurrence of creaming. Whereas emulsions that showed 0%T and no decrease in %BS at the bottom of glass cell (left side in the figure), from emulsions prepared with dispersions SMP:P-RT ratios 15:1 and 5:1 (Figure S1e and g, respectively), no creaming was observed since a decrease in %BS at the bottom of cell glass (0–25 mm) would indicate a decrease in droplets concentration caused by creaming. Therefore, for the emulsions prepared with dispersions SMP:P-RT ratios 15:1 and 5:1 (Figure S1e and g, respectively), higher stability against creaming was possibly related to the increasing apparent viscosity (Fig. 5a). The creaming was more intense for samples prepared with the dispersions at lower P concentration, SMP:P-HT ratio 1:0, 25:1 and 15:1 (Figure S1b, d and f), which could be attributed to their low viscosity values (Fig. 5a). However, the emulsion prepared with dispersion SMP:P-HT ratio 5:1 also presented low viscosity value, showed reduced creaming, which may be attributed to the smaller droplet size of the emulsion (Fig. 4b).

Variation of BS% in the middle of the glass cell (25–55 mm) is correlated with flocculation and/or coalescence since BS% decreasing indicates droplet size increase. The emulsion with dispersion SMP:P-RT ratio 1:0 presented the highest delta BS% showing that



RT	Mode (μm)
SMP:P 1:0	26.86 ± 2.02[a]
SMP:P 25:1	17.04 ± 0.99[b]
SMP:P 15:1	18.53 ± 0.06[b]
SMP:P 5:1	16.24 ± 0.02[b]

HT	Mode (μm)
SMP:P 1:0	21.41 ± 0.05[a,b]
SMP:P 25:1	19.48 ± 1.49[b,c]
SMP:P 15:1	17.80 ± 1.28[c,d]
SMP:P 5:1	14.90 ± 1.11[d]

Figure 4 Profiles of droplet size distribution and mode (μm) of oil in water (o/w) emulsion prepared with biopolymer dispersions of sunflower meal (SM) and pectin (P), SMP:P ratio 1:0, 25:1, 15:1 and 5:1 (w/w), where SMP corresponds to SM proteins, at (a) room temperature (SMP:P-RT) and (b) previously heat treated (SMP:P-HT).

flocculation and/or coalescence occurred (Figure S1a). The emulsions were prepared with dispersions close to the zero-point charge of SM, which may have favoured flocculation and/or coalescence due to hydrophobic attraction between proteins adsorbed on the droplets, since the electrostatic repulsion is weak to overcome attractive forces (McClements, 2004). However, the emulsion prepared with dispersion of SMP:P-HT ratio 1:0 showed no flocculation and/or coalescence (Figure S1b). According to Mitidieri & Wagner (2002), this effect can be attributed to the increased surface hydrophobicity and denatured proteins improved the stability of the interfacial film due to heat treatment.

Electrostatic interactions between SMP and P at pH 3.7 reduced flocculation or coalescence rate for emulsions prepared with dispersions of SMP:P ratios of 25:1, 15:1 and 5:1 at RT or HT (Figure S1) due to the net negative charge of electrostatic complexes (Fig. 1). Protein–polysaccharide electrostatic complexes reduce van der Waals attraction forces and increase steric and electrostatic repulsive forces between droplets, improving the emulsion stability (Xu *et al.,* 2017). In

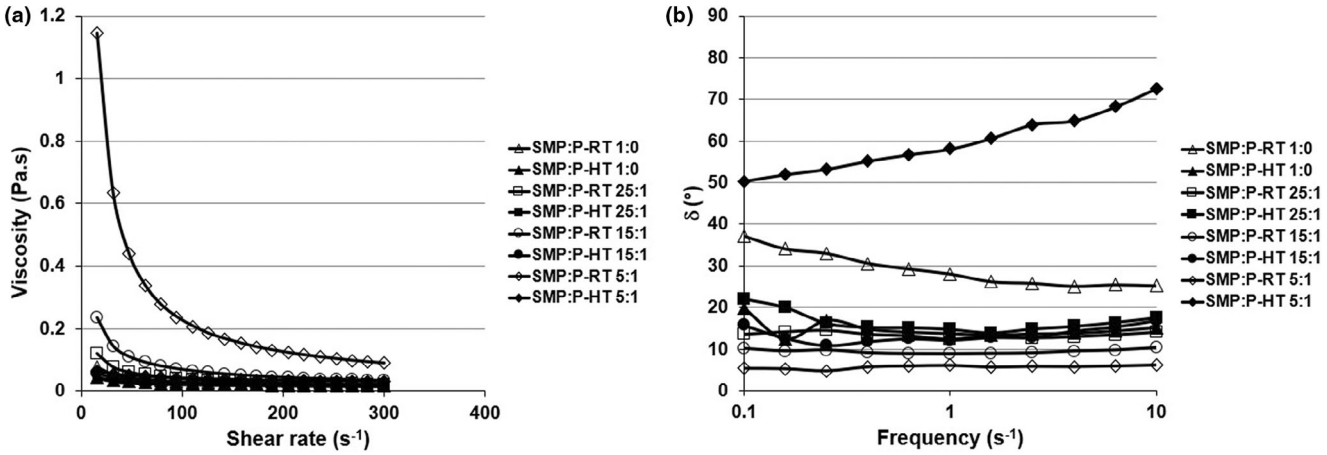


Figure 5 (a) Apparent viscosity of emulsions prepared with sunflower meal (SM) and pectin (P) dispersions, SMP:P ratio 1:0, 25:1, 15:1 and 5:1 (w/w), where SMP corresponds to SM proteins, at room temperature (SMP:P-RT, open symbol) and previously heat treated (SMP:P-HT, filled symbol) and (b) loss angle (δ) as a function of frequency of emulsions stabilised by SMP:P ratio 1:0, 25:1, 15:1 and 5:1 (w/w) dispersions, at room temperature (SMP:P-RT, open symbol) and previously heat treated (SMP:P-HT, filled symbol).

addition, with the increase in P concentration (SMP:P-RT ratio 1:0 to 5:1), the emulsions presented gel-like structure (Fig. 5) indicating that the interfacial film presents greater rigidity, contributing to a higher emulsion stability (Palazolo *et al.,* 2005).

Conclusions

It was observed that the biopolymer interactions and structural characteristics of SMP and P dispersions at pH 3.7 depend on the ratio between them and the heat treatment. Rheological properties and microstructure of the mixed colloidal dispersions exerted influence on the emulsion stability by net negative charge of electrostatic complexes and gel-like structure. The addition of pectin increased the apparent viscosity and gel-like behaviour ($G' > G''$) for biopolymer dispersions prepared at RT and for their emulsions. The microstructure of SMP:P ratio 5:1 dispersion showed protein attached to pectin and individual pectin chain in the continuous phase. In this condition, the emulsion presented stability related to creaming and flocculation and/or coalescence. Whereas dispersions prepared at 98 °C showed low apparent viscosity and increasing pectin concentration up to SMP:P ratio 5:1, dispersion presented spheroid particles and emulsion behaved as liquid-like material. The lower creaming rate was due to smaller droplet size.

Therefore, controlling the structure and rheological properties of biopolymer dispersions by P addition and HT, SM may be applied as ingredient in various food products development. SMP:P electrostatic complex may be applied in acidic food products, such as, dressings, sauces, dips and fermented beverages. Whereas SMP:P-HT might be used in non-acidic food products since heat treatment improves electrostatic complex stability regarding pH variations.

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Author contributions

Raquel Carolina Giarola: Conceptualization (equal); data curation (equal); formal analysis (equal); investigation (equal); methodology (equal); writing – original draft (equal); writing – review and editing (equal). **Roseli Aparecida Ferrari:** Conceptualization (supporting); funding acquisition (lead); project administration

(lead); writing – review and editing (equal). **Ana Clara Troya Raineri Fiocco:** Conceptualization (equal); data curation (equal); formal analysis (equal); methodology (equal); writing – review and editing (equal). **Flavia Maria Netto:** Conceptualization (supporting); data curation (supporting); methodology (supporting); writing – review and editing (equal). **Rosiane Lopes da Cunha:** Conceptualization (supporting); data curation (supporting); methodology (supporting); writing – review and editing (equal). **Elizabeth Harumi Nabeshima:** Conceptualization (supporting); methodology (supporting); resources (equal); writing – review and editing (supporting). **Peter Eisner:** Conceptualization (supporting); data curation (supporting); methodology (supporting); writing – review and editing (supporting). **Isabel Muranyi:** Data curation (supporting); formal analysis (supporting); methodology (supporting); writing – review and editing (supporting). **Mitie Sonia Sadahira:** Conceptualization (lead); data curation (lead); formal analysis (lead); investigation (lead); methodology (lead); supervision (lead); visualization (lead); writing – original draft (lead).

Conflict of interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: The investigations of the sunflower meal presented in the present study were part of the bilateral Brazil–Germany cooperation project SunflowerProtein funded by BMBF – Bioeconomy International, Project no. 031A281A. In the meantime, the patent for the process for the production of the sunflower meal has been granted in 2019: PROCESS FOR THE PRODUCTION OF PROTEIN PREPARATIONS FROM SUNFLOWER SEEDS, EP2400859. The fact that the sunflower meal was patented had no influence on the results presented.

Ethics approval

Ethics approval was not required for this research.

Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Figure S1. Transmission (%T) and backscattering (%BS) profile vs. glass cell height (mm), from bottom (left side) to top of glass cell (right side), and time (h) of emulsions prepared with sunflower meal (SM) and pectin (P) dispersions, SMP:P ratio 1:0, 25:1, 15:1 and 5:1 (w/w), where SMP corresponds to SM proteins, at room temperature (SMP:P-RT) and previously heat-treated (SMP:P-HT).

Table S1. Power law index n, K and R^2 and apparent viscosity at 15.8 s^{-1} of sunflower meal (SM) and pectin (P) dispersions, SMP:P ratios 1:0, 0:1, 25:1, 15:1 and 5:1 (w/w), prepared at room temperature (RT) and heat-treated (HT).