Particle Size Distribution and Viscoelastic Behavior of French Dressing Containing Two Types of Commercial Waxy Maize Starches

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ABSTRACT: Emulsion stability, viscoelastic rheological properties and particle size distribution of French dressing samples formulated with two types of commercial modified starch have been investigated. The product's usual thickening agents, xanthan and guar gum, were 100% substituted by (2, 2.2%) of acetylated distarch adipate (ADA) and hydroxypropylated distarch phosphate (HDP) from waxy maize starch. The samples with 2% hydroxypropylated distarch phosphate starch showed the minimum emulsion stability (<95% after three months of storage) and were eliminated from further studies and the other three samples along with the control were selected for particle size distributions and dynamic rheological properties analysis. The sample formulated with 2.2% of HDP starch showed a better storage stability (higher zero shear viscosity), deformability (γ_{LVE}) and greater texture strength (G'₀) due to more structural entanglement and intermolecular hydrogen bonds as compared to the other type of commercial starch and control. In particle size distribution curve of this sample a broad and distinguished peak with higher percentage of population ranging from 10 to 100 µm was observed which might be due to the higher water absorption and more swollen/degraded starch granules. The area-based average diameter (d_{3,2}) of the particles confirmed more similarity between this sample and control.

Keywords: Particle Size Distribution, Salad Dressing, Viscoelastic Rheological Properties, Waxy Maize Starch.

Introduction

Salad dressings are classified as o/w emulsions with different formulations and commercial names such French. as Thousand island, Sezar and Italian dressings (Franco, 1997; Mandala, 2004). One of the main group of ingredients in this product usually act as stabilizers/texture that improvers are hydrocolloids such as xanthan, guar gum and different types of starches (Mandala, 2004; Arocas, 2009). Native starches are rarely used in food because of weak processing systems functional properties especially high

tendency to syneresis. Todays several physical, chemical and/or enzymatic methods are used to modify the native starch to meet the viscosity requirements and improved heat/acid stability for food industry (Chui and solark, 2009; Wurzburg, 1986; Rosalin, 2002). Hydroxy propylation, acetylation and chemical cross-linking are the common modifications results in better stability against low pH, high temperature and shear stresses (Ahmt, 2004; Abass, 2010). The main criteria to consider when selecting an optimum hydrocolloid for salad dressing are good tolerance against high shear stresses and acidic conditions. Many studies have focussed on the rheological

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properties of different starch-based systems subjected to different mixture of hydrocolloids, processing shear stresses and thermal conditions. In 1990, Muntoz, et al, studied the relationship between dynamic viscoelastic properties and the specific formulations of some commercial salad dressings. Franco et al. (1997) reported that the processing parameters might exert an important influence on the rheology and stability of salad dressings emulsions. Thebaudin et al. (1998) investigated the effect of thermal treatment on the textural characteristics of a béchamel sauce prepared with different starches. Influence of thermal treatment on the flow characteristics of starch-based food emulsions formulated with waxy maize starch was studied by Martinez et al. in 2003. Dolz et al. (2007) studied the effects of substituting starch with other gums such as xanthan and locust bean gum. The effect of emulsifier type and concentration of xanthan gum on physical properties of low fat sauces has been evaluated by Bortnowska et al. (2009). A complete review on the functional roles of different hydrocolloids in sauces and dressings has been introduced by Badrie et al. (2010). In all of the previous researches, starch combined with non-starch thickeners has been used to provide structural, textural attributes in emulsion-type food systems (Mandala, 2012).

The main objective of this study was to develop a new formulation for French dressing by 100% substitution of other hydrocolloids with different amounts of commercial modified waxy maize starches, as a low cost stabilizers and texture improver agents and investigate their effects on the emulsion stability, viscoelastic rheological properties and particle size distribution of the final product.

Materials and Methods

Acetylated waxy maize di-starch adipate (cold swelling, Instant Clear JEL-SD, National starch, USA, E1422) and hydroxypropylated waxy maize di-starch phosphate (cold swelling, Ultratex-SR. E1442) National starch. USA. were employed the specific physical with characteristics as shown in Table 1. Xanthan gum (GRINDSTED[®] xanthan 200, Danisco, Denmark) and guar gum (GRINDSTED[®] GUAR 250, Danisco, Denmark) were also used in the formulations. Other ingredients used in the French dressing formulations consisted of soybean oil, egg yolk, vinegar, sugar, red pepper powder, garlic powder, sodium benzoate, potassium sorbate, citric acid, mustard powder, salt and tomato paste were provided by the R&D center of the Behrouz Food Industries, Tehran, Iran.

- Preparation of French dressing samples

Five samples of French dressing were prepared with the usual ingredients consisted of fresh egg yolk (10%), vinegar (8%), soybean oil (38%), salt (2%), mustard powder (0.4%), sugar (6%), tomato paste (5%), red pepper (0.08%), citric acid (0.1%), garlic powder (0.04%), sodium benzoate (0.063%) and potassium sorbate (0.012%). Acetylated di-starch adipate and hydroxypropylated di-starch phosphate were used in the samples according to the Table 2.

Table 1. Physical properties of commercial waxy maize starches (Yaghoti moghadam et al., 2012)

Type of Starch	Instant Clear JEL-SD	Ultratex-SR
Apparent Viscosity(Pa.s)	1.52	0.51
Water Holding Capacity(g/g)	22.26	24.29
Swelling Power (g/g)	9.16	8.22

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Table 2. Code of	samples prepared	with different types/amounts	of modified starches

Samples	Modified starch	Type of mofification	percentage of modified starch
R1	Ultra Tex-SR	Hydroxypropylated di-starch phosphate	2%
R2	Ultra Tex-SR	Hydroxypropylated di-starch phosphate	2.2%
R3	Instant clearjel-SD	Acetylated di-starch adipate	2%
R4	Instant clearjel-SD	Acetylated di-starch adipate	2.2%

Control sample was prepared with xanthan and guar gum instead of starch. All of the samples were sealed in glass jars (200 ml) and kept in the refrigerator (4°C) until required for further analysis concerned with the following physical and rheological tests.

- Emulsion stability

The stability of the French dressing samples was assessed according to the method of Masken *et al.* (2000), in triplicate, after 1 and 3 months of storage at 4° C.

- Steady flow behavior

Rheological measurements were studied by rheometer (Paar Physica MCR 300, Anton Paar GmbH, Austria) with a serrated surface parallel plates (25 mm diameter, a gap distance of 1 mm) at 25° C. Flow behavior tests were accomplished at shear rates in the range of $10^{-4} - 300 \text{ s}^{-1}$. In order to characterize the steady flow behavior, the experimental data were fitted to the Carreau model, equation 1 (Dolz, 2007) and the rheological parameters were calculated by US 200 V 2.21 software:

$$\eta_{app} = \eta_0 / [I + (\lambda \gamma)^2]^c \qquad \text{Eq (1)}$$

 η_{app} : Apparent viscosity γ : Shear rate η_0 : Zero shear viscosity (pa.s) λ : Characteristic time (s) c: (n-1)/2

- Viscoelastic behavior

Linear viscoelastic strain (γ_{LVE}) and structural strength (G'max) of the French dressing samples were determined through

strain amplitude sweep tests (0.01-100%) at the constant frequency of 1 *Hz*. Frequency sweep tests were performed at constant strain ($\gamma < \gamma_{LVE}$), between 0.1 to 100 *Hz* (Difts, 2005). The results were then fitted to the Bohlin model (equation 2):

$$G' = a\omega^b$$
 Eq(2)

G': Storage moduli (pa)

a: Proportional coefficient (-) ω : Angular frequency (1/s) b: Coordination number ()

b: Coordination number (-)

Finally the suitability of Cox- Merz rule was examined by studying the products behavior in the two types of flow curves: apparent viscosity versus shear rate (η_{app} / γ') and complex viscosity versus frequency (η^* / ω).

- Particle size analysis

The particle size distribution of French dressing samples have been studied by a Malvern particle size analyzer (Mastersizer, 2000) according to the procedure described by Arancibia *et al.* 2013.

Results and Discussion

- Emulsion stability

The effect of different types/amounts of modified starches on the emulsion stability of French dressing samples during storage for 1 and 3 months has been shown in Table3.

- Steady-states rheological behavior

Flow behavior curves (apparent viscosity/shear rate) of salad dressing

samples containing different types and amounts of modified starches are shown in Figure 1. All of the samples exhibited a similar shear thinning behavior that is the dramatic shear-induced result of the structural break down and deflocculating of oil droplets aggregates (Moros et al., 2002). The rheological parameters are determined after fitting the experimental data to the simplified Carreau model that is usually considered as the best-fit model for starch containing dispersions (Chamberlain & Rao, 1999; Zimeri & Kokini, 2003; Dolz, 2007), gels (Martinez, et al., 2003), and emulsions (Cabeza et al., 2002; Moros et al., 2002;

Riscardo *et al.*, 2003). It might be emphasized that in all of the previous investigations, different types of starches were used along with other hydrocolloids but in the present research, in spite of the fact that the only thickener is starch, the results showed a good fit (R \geq 0.98) to carreau model. No significant difference has been observed in the slope of power law region of the samples (Table 4). This means that the apparent viscosity of starch- based emulsion is decreased by shear rate in the same rate because of the weak structural bonds (Martinez *et al.*, 2003).

Table 3. Emulsion stability of French dressing samples during storage

Sample	Emulsion stability		
	1 month	3 months	
Control	97.8 ^{cd}	96.80 ^a	
R1	96.02 ^{bc}	1	
R2	97.30 ^{bd}	96.30 ^a	
R3	98.10 ^{cd}	97.05 ^b	
R4	98.33°	98.16 ^b	

¹ After 3 months the sample was completely separated.

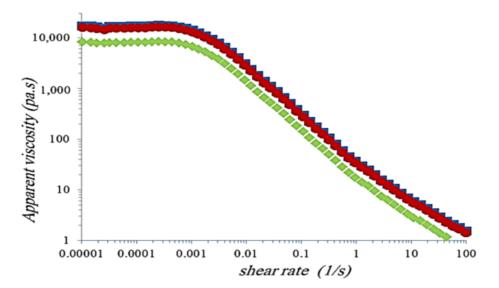


Fig.1. Flow behavior curves (apparent viscosity/shear rate) of French dressing samples.Control \blacklozenge , R2 \blacksquare , R3 \blacktriangle , and R4 \blacklozenge (*a*) 25°C.

Another parameter obtained from this model is zero shear viscosity (η_0) , the viscosity in very low shear rates (10^{-4} s^{-1}) which might be related to the storability of the samples. According to the Table4, η_0 was varied in the range of $(0.8-1.7) \times 10^4$ pas with significantly higher values for starch containing samples as compared to the control. Therefore 100% substitution of xanthan and guar gum in control with starch might result in a more viscose emulsion with better stability during storage. However η_0 value might also depend on the type and quantity of the thickening agent (Dolz, 2007) and higher amounts (2.2%) of hydroxypropylated di-starch phosphate waxy maize starch resulted in a desirable increase in this factor for R2. Martinez et al. (2003), showed that in starch-based emulsions, swelling of starch granules and formation of the intermolecular bonds might be responsible for higher viscosity in lower shear rates

- Viscoelastic behavior

Viscoelastic tests are usually used to predict emulsions structural changes during processing and storage (Lai & Lin, 2004). Table 5 shows the linear viscoelastic limit strain (γ_{LVE}) and the storage modulus at γ_{LVE}

 (G'_0) for different French dressing samples and the control. γ_{LVE} and G'₀ are usually considered as the indices of deformability structural strength, respectively and (Mancini, Among the starch 2002). containing samples, R2 had similar deformability (γ_{LVE}), but stronger structure as compared to the control (Table 5) and it means that this type of modified waxy maize starches acts more successfully as thickening agent than xanthan and guar gum. Arocas et (2009)investigated al. the hydroxypropylated and acetylated modified starch on the formulated white sauce and reported the same result. It should be noticed that the amount of oil used in O/W emulsions is considered a key factor influencing deformability the and consistency of the final product and in the present research a good texture has been achieved by using modified starch along with a suitable amount of oil.

Values of damping factor (tan δ) also confirmed this point that different types of modified starch might strengthen the continuous phase of the emulsion samples and improve its solid like behavior that in turn results in lower tan δ , in comparison to the control (Table 5).

Table 4. Carreau model parameters of French dressing samples^{1,2}

Samples	zero-shear viscosity (η_0 , pas)	characteristic time (λ, s)	slope of the power law region (p, -)	R ²
Control	8050 ^a	756 ^a	0.43 ^a	0.99
R2	17810 ^c	765 ^a	0.45 ^a	0.98
R3	16446 ^b	761 ^a	0.45^{a}	0.99
R4	16102 ^b	760^{a}	0.56^{a}	0.99

¹ Results are expressed as mean values. ² Data with similar superscript letters represent no significant difference (p<0.05).

Table 5. Linear viscoelasticity limit strain (γ_{LVE} ,%) and G'₀ for French dressing samples^{1,2}

Samples	γ _{LVE} (%)	G' ₀ (pa)	tanð
Control	0.48 ^a	145.11 ^a	0.36 ^a
R2	0.40 ^a	344.43°	0.17 ^b
R3	0.35 ^b	239.41 ^b	0.16 ^b
R4	0.33 ^b	312.52 ^b	0.16 ^b

¹ Results are expressed as mean values. ² Data with similar superscript letters represent no significant difference (p<0.05).

According to the results of frequency sweep tests (Figure 2), all of the samples showed a solid viscoelastic behavior $(G^2 \geq G^{"})$ and frequency dependent and might be classified as weak gels. This behavior has been already reported for salad dressings (Lai & Lin, 2004; Difits, 2005; Dolz, 2006, 2007; Zhen Ma, 2013). The storage and loss modulus of the samples containing modified starches were higher than the control that was formulated with xanthan & guar gum (Figure 2). This might be due to the more molecular entanglement and intermolecular hydrogen bonds that are the most important factors having effects on G' and G" in emulsions with modified starches than other stabilizers. This point might be more clearly observed in the results of tan δ (G"/G') in Figure 3. Since $\tan \delta$ was in the range of 0.1 - 1, all of the samples, might be considered between the true gels and a concentrated biopolymer and this is а known characteristic structure for salad dressings

(Dolz et al., 2006).

The frequency dependence of the storage modulus (G') for French dressing samples might be described by the power law model. In this model, "b" and "a" values indicate the level of interactions between polymer chains and its magnitude, respectively (Peressini, *et al*, 1998). According to Table 6, the highest "a" value was related to the sample containing Ultratex-SR, where no significant difference was observed among the "b" values. These results confirmed that the interactions between starch chains are stronger than xanthan and guar gum as stabilizers and might provide better stability in the final product.

The comparison between complex viscosity (small deformation) and steady shear viscosity (large deformation) is shown in Figure 4. According to Cox/Merz rule, for many polymers and solutions, the shear viscosity function (η/γ) from rotational tests, and the function of complex viscosity

Table 6. Power law parameters of G' for French dressing samples^{1,2}

Samples	k	n	D _[3,2]	
Control	57.54 ^a	0.61 ^a	13.02 ^a	
R2	170.18 ^c	0.49^{b}	15.53 ^a	
R3	131.82 ^b	0.52^{b}	9.63 ^b	
R4	147.91 ^b	0.47 ^b	9.08 ^b	

^TResults are expressed as mean values. ² Data with similar superscript letters represent no significant difference (p<0.05).

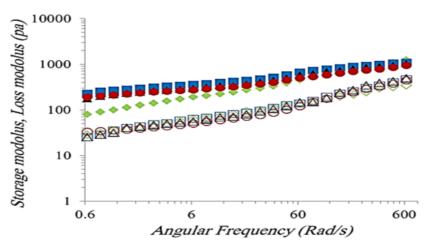


Fig. 2. Dynamic mechanical curves of salad dressing samples: Control \blacklozenge , R2 \blacksquare , R3 \blacktriangle , and R4 \bullet measured at 0.3% strain and 25°C. Full symbols represent storage modulus (G') and empty symbols represent loss modulus (G').

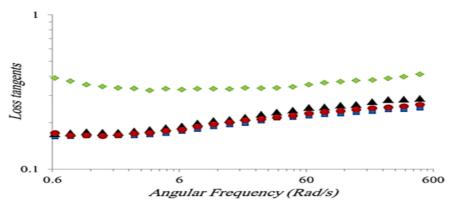


Fig. 3. Tan δ of salad dressing samples: Control \blacklozenge , R2 \blacksquare , R3 \blacktriangle , and R4 \bigcirc

 (η^*/ω) from oscillatory tests, display an identical curve over a wide range, when represented in the same diagram (Mezger, 2002). But for dispersion systems, where physical and/or chemical interactions might occur between different ingredients and affect the rheological properties, some deviations might be observed from this rule. In Figure 4, this deviation has been shown, therefore the shear viscosity of salad dressings are lower than the complex viscosity. This means that higher shear rates might cause more severe shear- thining behavior in this type of dispersion/ emulsion food products which is typical а phenomenon for weak gels (Worrasinchai et al., 2006).

- Particle size distribution

The particle size distributions of different French dressing samples are shown in Figure5 (a,b,c). In control (curve a) two main peaks: one at (1-10) μ m and the other at (100-1000) μ m are observed. The bigger peak (8.93 μ m) is related to the small, free oil droplets and those stabilized by a layer of egg yolk proteins in normal dressing (Muller *et al.*, 1998; Worrasinchai *et al.*, 2006; Rayner *et al.*, 2012; Arancibia *et al.*, 2013) and the smaller peak (200-500 μ m) comes from the coarse particles of tomato paste in the dressing formulation (Bayod, 2008; Juszczak, 2013). Whereas, in the curves of samples formulated by modified starch (b, c), three main peak has been appeared, the first and third ones are somewhat similar to the control and the second peak (10-100 μ m) could be considered to represent the dispersed swollen starch granules. Juszczak et al. (2013) have investigated the structural properties of ketchup and reported that the particle size of granules of acetylated distarch adipate (ADA) corn starch in this suspension- type product is in the range of 50-100 µm. The effect of starch on the particle size distribution of o/w emulsion model systems has been studied bv Arancibia 2013 and a bimodal particle size curve was observed in all of the samples. However It was emphasized that the water holding capacity (WHC) of the starch thickener and its concentration in continuous phase were the main parameters affecting the particle size distribution of the final product (Arancibia, 2013). This point might be obviously distinguished by comparing the second peak in the Figure 5, curves b and c. It was previously mentioned that WHC of hydroxypropylated di-starch phosphate waxy maize starch (Ultratex-SR) is about 3 times greater than that of acetylated di-starch adipate waxy maize starch (Instant Clear Jel-SD) as presented in Table 2. Therefore the former starch granules might have been much more swollen and partially degraded into smaller particles and have created a broad peak with higher percentage of population ranging from 10 to 100 µm

(Heyman *et al.*, 2014). As the area-based average diameter $(d_{3,2})$ of particles is usually considered to evaluate the textural properties of the final product (Bayod, 2008), it might be concluded that R2, the sample formulated

with 2.2% Ultratex-SR has a texture quite similar to the control (Table 6).

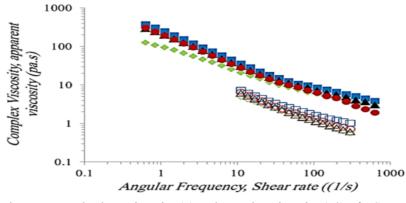


Fig. 4. Comparison between steady shear viscosity (η) and complex viscosity (η^*) of : Control \blacklozenge , R2 \blacksquare , R3 \blacktriangle , and R4 \bullet French dressings samples measured at 25 °C. Full symbols represent complex viscosity and empty symbols represent steady shear viscosity.

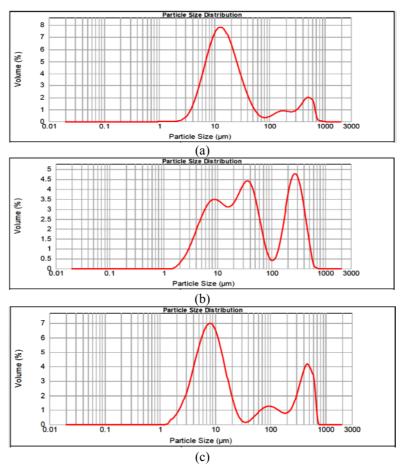


Fig. 5. Particle size distribution of French dressing samples a) control b,c) thickened with hydroxypropylated distarch phosphate and acetylated distarch adipate waxy maize starch

Conclusion

A new formulation for a high consuming condiment, French dressing, has been developed by using 2.2% of a low cost thickening agent instead of xanthan and guar gum. As there are differences between the nutritional values of hydroxypropylated distarch phosphate and gums, it might be essential to evaluate the health benefits of this new formulated food product in the future studies.

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