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EFFECT OF MODIFIED STARCH/ NON-STARCH THICKENER COMBINATION ON CONSISTENCY, STABILITY AND RHEOLOGICAL PROPERTIES OF TOMATO KETCHUP

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ABSTRACT

Ketchup is one of the most popular tomato products on the world market and requires limited equipment and simple processing. Thickeners are used in the manufacturing process due to their ability to act on the viscosity, affect the consistency and prevent the ketchup from delaminating.

The effect of two modified starches in combination with a non-starch thickener (guar gum, xanthan gum and carrageenan) was investigated on the consistency, stability and rheological properties of tomato ketchup. A two-way ANOVA was performed to evaluate the effects of starch and non-starch thickener on structural mechanical properties and Bostwick consistency of ketchup.

All samples appeared to be non-Newtonian fluids and their viscosity and variation were close. Ketchup samples showed the highest shear stress values with 0.2% carrageenan with 3.4% modified potato starch, while the lowest were shown for samples with 0.1% guar gum. The highest consistency values determined by the Bostwick method of ketchups were reported for the combination of 3.4% modified potato starch and 0.1% guar gum, and the lowest for 3.8% modified waxy corn starch and 0 .2% carrageenan. During the analysis of the obtained samples, the serum-separated liquid was detected in ketchup with only modified potato starch, in combination with guar gum, in an amount of 0.1%. Based on these results, the combination of modified waxy corn starch and 0.2% carrageenan was the most suitable to be used for the production of tomato ketchup, with the aim of creating a more sustainable product.

1. Introduction

The production of tomatoes (Lycopersicon esculentum) is one of the first in the world among vegetables. Tomatoes can be eaten raw, but due to their perishable nature, they are processed (Quinet *et al*., 2019). Much of the world's tomato crop is processed into tomato paste, which is subsequently used as an ingredient in many food products, mainly soups, sauces and ketchup (Cammarano *et al*., 2022; Roccotiello *et al*., 2022). Tomato ketchup is an easy-to-use and low-calorie product made from concentrated tomato paste with spices, salt, sugar and vinegar with or without starch, onion and garlic and contains no less than 12% tomato solids (Alqahtani, 2020; Mohamed *et al*., 2020). Tomatoes have been used to modify the taste and/or aroma of certain foods and culinary preparations, with consistency and colour widely appreciated by consumers (Ahouagi *et* *al.,* 2020). Although tomato ketchup is the most commonly used snack condiment in homes and restaurants, its nutritional value and biofunctional properties are limited to the nutrients and bioactive compounds present in tomatoes and their stability during and after processing (Prakash *et al.*, 2016; Ahouagi *et al.*, 2021; Szabo, 2022). Tomatoes, as the main ingredient in ketchup, are recognized as a source of carotenoids (lycopene), a very important class of bioactive compounds, particularly known for their anti-inflammatory properties and supporting prostate health (Salehi *et al.*, 2019; Przybylska, 2020; Coelho *et al.*, 2023). There are many types of ketchup in the market, such as baby ketchup, fine, spicy, ketchup with different types of flavours, etc. These ketchup differ mainly in the content of the main ingredient, i.e. the tomatoes and spices used, as well as stabilizers (modified starch, pectin), which are often widely used (Fritsch *et al.*, 2017; Himashree *et al.*, 2022). Important characteristics of this type of product are its stability, consistency and rheological properties. Ketchup is a thin liquid with a yield point. It also exhibits thixotropic and viscoelastic properties (Torbica *et al.*, 2016; Li *et al.*, 2017; Shokraneh *et al.*, 2023).

The rheological properties of ketchup are mainly influenced by the rheological characteristics of tomato concentrate (Anamaria and Giani, 2019; Stanciu *et al.*, 2020; Gao *et al.*, 2021). The volume fraction of solids is the most important parameter affecting the rheological properties of tomato concentrate and ketchup (Wang *et al.*, 2018; Gao *et al.*, 2021). The viscosity is one of the main quality aspects that must be considered to determine the overall quality and consumer acceptability of many tomato products. The degree of ripeness, processing temperature, solids content, particle size and number of particle interactions play a role in determining the viscosity of tomato products (Shatta *et al.*, 2017; Jayathunge *et al.*, 2018). The consistency is related to non-Newtonian or semi-solid liquids (sauces, purees and pastes) with suspended particles and long-chain soluble molecules and is practically measured by product distribution or flow (Kumbar *et al.*, 2019; Pirsa and Hafezi, 2023). The consistency and rheological properties of ketchup depend not only on the amount of tomato paste used and its rheological characteristics but also on the type and amount of thickeners added (Torbica *et al.*, 2016; Diantom *et al.*, 2017; Thanh-Blicharz and Lewandowicz, 2020; Himashree *et al.*, 2022). Starch is a functional and commonly used food component. However, in its natural form, it shows low rheological stability and low resistance to mechanical, thermal and chemical agents. Furthermore, it undergoes retrogradation and syneresis phenomena, which limit the use of natural starch in many food products. To improve certain physicochemical properties of natural starch, it can be modified by chemical, physical and/or enzymatic methods or combinations thereof. The resulting starch preparations exhibit various functional properties and are used as gelling, thickening, stabilizing and fillers in food production (Zia-ud-Din *et al.*, 2017; Liu and Xu, 2019; Cui *et al.*, 2022; Obadi *et al.*, 2023). Much research has been done to obtain the properties required for a particular application using mixtures of starch and hydrocolloids. Previous research has reported that mixtures of starch and hydrocolloids have been used as thickeners and/or stabilizers to control water mobility, facilitate processing, and improve stability in food systems (Li and Nie, 2016; Mahmood *et al.* 2017; Cai *et al.*, 2020). The most commonly used thickeners are polysaccharide hydrocolloids such as: guar gum, xanthan, tragacanth, pectins and sodium alginate (Li and Nie, 2016; Pirsa and Hafezi, 2023).

This work aimed to evaluate the effect of two types of commercial modified starches of different origins in combination with different types of hydrocolloids on the consistency, stability and rheological properties of ketchup. Rheological properties of tomato ketchup such as viscosity and flow curves were analyzed for 24 tomato ketchup combinations.

2. Materials and methods

2.1. Materials

The following modified starches and hydrocolloids were used: Acetylated distarch adipate from waxy maize starch (Resistamyl 341), Tate & Lyle (R); Chemically modified potato starch (cross-linked esterified distarch adipate) (Adamil 2075 starch), Global Ingredient MSK (A), Gum guar (GG), Biovegan, German, Gum Xanthan (GX), ZoyaBG, Chaina, Carrageenan Iota-type pure semi-refined carrageenan whose composition has natural high polymeric hydrocolloid extracted from red algae (Eucheuma Denticulatum) (TS-200), Tacara SDN BHD, Malaysia (TS) and the other materials were from the local market.

2.2. Methods

2.2.1. Preparation of ketchup

Ketchup was made by mixing all the ingredients: sugar, starch, nonstarch thickener, salt, citric acid, tomato concentrate (36°Brix) and water. The mixture was homogenized and then heated at 75°C for about 7 minutes. Samples (25-26°Brix; pH 3.4-3.8) were allowed to cool and stood for at least 24 hours before analyses. There were made 24 samples with two types of modified starch (R and A) in two concentrations (3.4% and 3.8%) in combination with nonstarch thickeners gum guar (GG), gum xanthan (GX) and carrageenan (TS) in two levels (0.1 and 0.2 %), shown in Table 1.

Table 1. Quantity and type of starch/hydrocolloid combinations used as thickeners in the ketchup composition

Starch/non-starch thickener, %	GG(0.1)	GG(0.2)	GX(0.1)	GX(0.2)	TS(0.1)	TS(0.2)
A(3.4)	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6
A(3.8)	Sample 7	Sample 8	Sample 9	Sample 10	Sample 11	Sample 12
R(3.4)	Sample 13	Sample 14	Sample 15	Sample 16	Sample 17	Sample 18
R(3.8)	Sample 19	Sample 20	Sample 21	Sample 22	Sample 23	Sample 24

2.2.2. Rheological measurements

All measurements were carried out at a constant temperature of 25 °C. Experimental results were modelled using a power law, also known as the Ostwald-de Waele model. Rheological characteristics were determined using a Rheotest-2 rotational viscometer (RHEOTEST Medingen GmbH, Medingen, Germany), within the shear rate range from 0.33 to 145.8 s^{-1}.

The dynamic viscosity (η, Pa.s) was calculated (Rao, 2014) using the formula:

$$\eta=\tau/D \qquad (1)$$

Where τ is the shear stress, Pa; D is the shear rate, s^{-1}.

The most used model for complex fluids is the famous power law dedicated principally to the Ostwald-de Waele model (Stanciu *et al.*, 2020), which is simply expressed as follows:

$$\tau=k.D^{n} \qquad (2)$$

Where K is the flow consistency index, Pa.s; n – is the flow behaviour index, D is the shear rate or the velocity gradient perpendicular to the plane of shear, s^{-1} and τ is the shear stress (Pa)

Determination of Bostvik consistency

Flow lengths (after 30 s) of the samples (at 20°C) were measured using a standard Bostwick consistometer (Operating instruction, Bostwick consistometer, Labomat). The results were obtained as the average values of three parallel measurements (McCarthy and McCarthy, 2009).

2.2.3. Determination of syneresis resistance

Following centrifugation (Hettich Zentrifugen EBA 200) for 15 min at 3000 min^{-1}, the separated liquid was quantified by weight and expressed as a percentage of the sample.

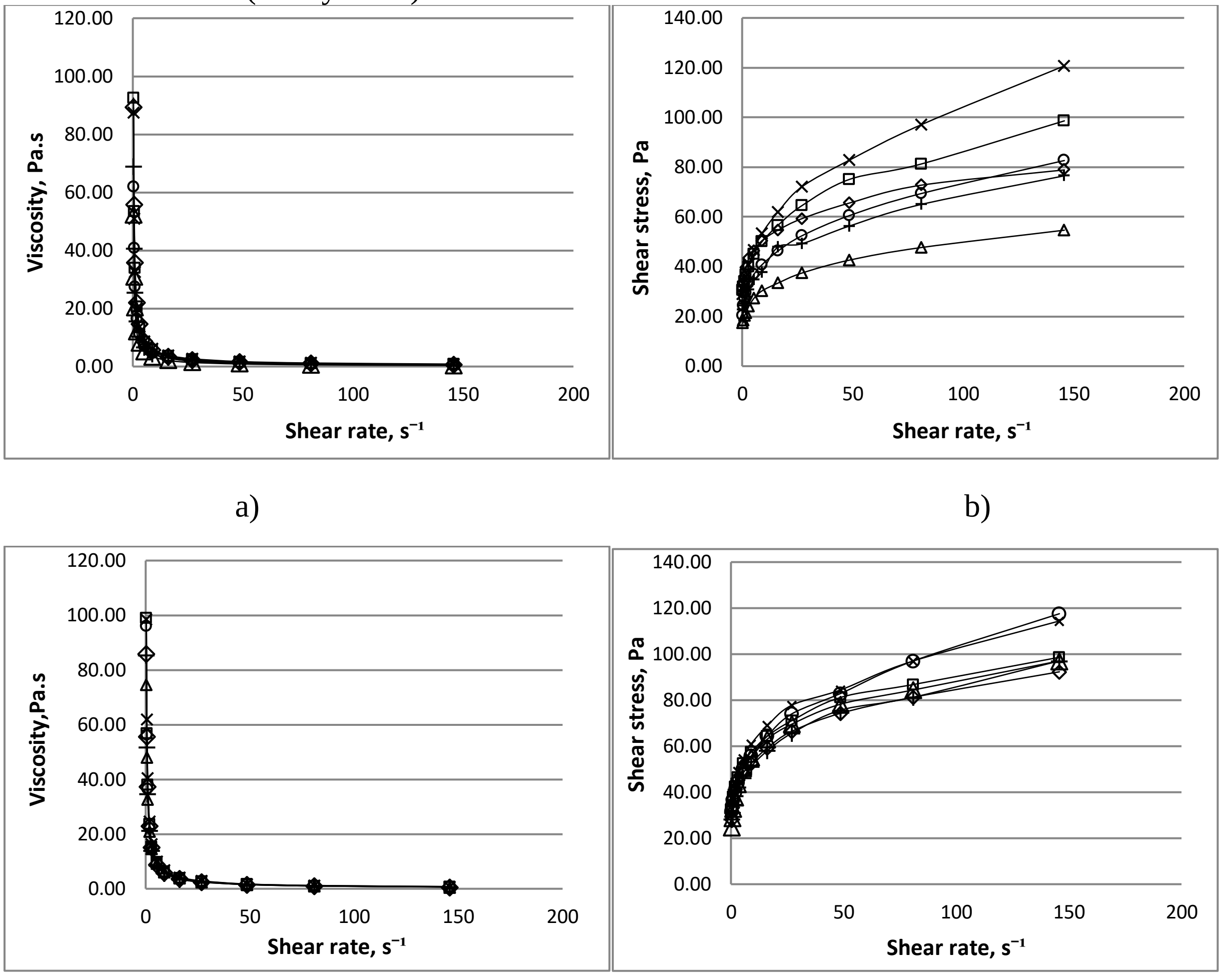
2.2.4. Statistical analysis

A two-way ANOVA was conducted to examine the effects on the ketchup properties of different starch/hydrocolloid combinations used as thickeners. The data are presented as means±standard deviation. The data were submitted for analysis of variance partitioned into components attributable to different sources of variation (starch, hydrocolloid, interaction starch-hydrocolloid). The chosen level of significance was $\alpha=0.05$. The post-hoc analysis was performed using Tukey's Honestly-Significant-Difference (Tukey HSD) test.

3. Results and discussions

3.1. Rheological properties

The most important factor determining the structural-mechanical properties and quality of tomato ketchup is its viscosity (Kumbár *et al.*, 2019). Flow characteristics are an important parameter for all food products and this is information that is relevant to the economic design of the equipment used and the food processing operations to be selected (Ahmed *et al.*, 2017). Viscosity as a function of the velocity gradient "D" is presented in Fig. 1 (a) and c) for the samples with 3.4% and 3.8% modified starch R, e) and g) for the samples with 3.4% and 3.8% modified starch A) and different types of hydrocolloids in the amount of 0.1% and 0.2%.

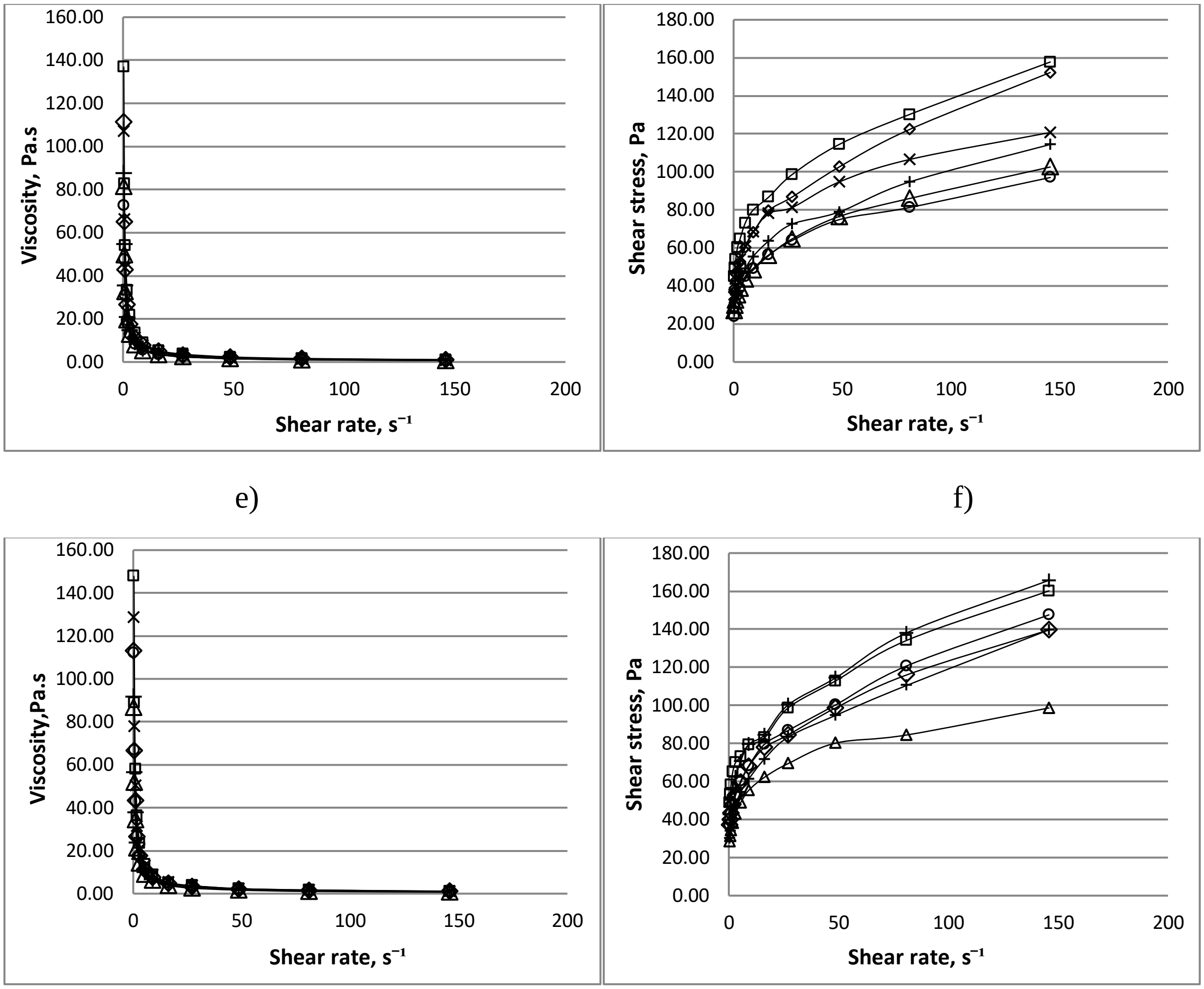


a)

b)

c)

d)



e) f)

g) h)

Figure 1. Viscosity and rheograms of ketchup with R 3.4% (a) and b)), R 3.8% (c) and d)) or A 3.4% (e) and f)), A 3.8% (g) and h)) and TS 0.1% (○), TS 0.2% (□), GG 0.1% (Δ),GG 0.2% (◊), GX 0.1% (+), GX 0.2% (X).

After obtaining the results, it was obvious that all the samples were non-Newtonian fluids. The shape of these curves indicates a shear-thinning non-Newtonian flow with a tendency toward yield stress. Non-Newtonian flow behavior of ketchup has also been observed by many authors (Berta *et al.*, 2016, Kumbár *et al.*, 2019). The highest shear stress values are shown for the ketchup sample with A(3.8%)+TS(0.2%), while the lowest is shown for the sample with A(3.4%)+GG(0.1%). These results correlate with consistency values, showing that ketchup thickened with A(3.4%)+GG(0.1%) showed the longest flow length (was the thinnest), while that with A(3.8%)+TS(0.2%) shows one of the shortest jet lengths (it was the thickest). The shear thinning behavior, i.e. a decrease in viscosity with increasing shear rate is a common phenomenon. In the case of ketchup, which is a product with a suspension structure, the shear thinning phenomenon results from the orientation of the tomato paste solids along the flow lines. The other factor affecting the viscosity of ketchup is the presence of swollen and partially gelatinized starch granules or their fragments. At higher shear rates, the individual starch granules may deform.

For samples containing modified waxy maize starch, the highest shear stress values were reported for samples with R(3.8%)+TS(0.2%), while the lowest were reported for samples with R(3.4%)+ GG(0.1%). The results have a similar dependence to those with modified potato starch.

Fig. 1 also presents the rheograms (b) and d) for the samples with 3.4% and 3.8% modified starch R, f) and h) for the samples with 3.4% and 3.8% modified starch A) are presented in Fig. 1. All samples show similar rheological behavior. From Fig. 1, the graphical correlation shows that in rheological terms the analyzed samples are non-ideal plastic bodies. The rheological behavior of the emulsions is typical of the Ostwald–de Waele models, which is evident from the coefficients of determination (R^2) obtained, ranging from 0.9651±0.0567 to 0.9994±0.0450. This model is widely used in the analysis of various food systems.

The consistency factor (k) and flow behavior index (n) obtained by fitting the power law and Ostwald–de Waele models to the experimental data for shear stress and shear rate as a function of temperature are shown in Table 2.

Table 2. Parameters of Ostwald–de Waele models for flow curves of the ketchup with modified starch/nonstarch thickener.

Sample	K	n	R^2	K	N	R^2
	Upward curve			**Downward curve**		
Sample 1	20.08±1.52	0.193±0.02	0.9932±0.0356	17.03±0.85a	0.230±0.12	0.9994±0.0450
Sample 2	34.14±2.34a	0.196±0.12	0.9809±0.0484	30.23±1.23bc	0.234±0.04	0.9975±0.0352
Sample 3	25.85±1.42b	0.204±0.03	0.9823±0.0235	23.04±2.35d	0.234±0.03	0.9947±0.0052
Sample 4	33.11±1.65ac	0.239±0.05	0.9827±0.1361	31.10±1.54bc	0.272±0.11	0.9989±0.0126
Sample 5	26.22±1.12b	0.218±0.03	0.9943±0.0089	19.47±3.25ae	0.275±0.06	0.9923±0.0225
Sample 6	35.71±2.03ad	0.158±0.11	0.9984±0.1230	22.54±1.25de	0.249±0.04	0.9991±0.0231
Sample 7	33.03±3.25ac	0.223±0.02	0.9953±0.0232	30.25±2.56bc	0.228±0.01	0.9969±0.0064
Sample 8	38.39±2.42dc	0.187±0.08	0.9973±0.0335	32.85±3.12b	0.217±0.05	0.9978±0.0356
Sample 9	34.15±2.36a	0.200±0.02	0.9958±0.0125	28.37±1.24cf	0.250±0.10	0.9984±0.0036
Sample 10	40.09±3.55e	0.200±0.05	0.9962±0.1136	33.97±2.13bg	0.242±0.02	0.9968±0.0187
Sample 11	36.14±2.32ad	0.185±0.10	0.9961±0.0356	26.28±1.25f	0.247±0.03	0.9983±0.0256
Sample 12	36.60±1.54ad	0.216±0.03	0.9851±0.0635	31.32±2.01bc	0.261±0.01	0.9851±0.0458
Sample 13	35.009±2.56ad	0.222±0.02	0.9903±0.0089	27.953±4.56cf	0.267±0.02	0.9929±0.0154
Sample 14	43.745±1.23f	0.200±0.02	0.9983±0.0023	36.381±3.24gh	0.223±0.06	0.9934±0.0025
Sample 15	30.847±2.41	0.224±0.01	0.9980±0.0129	26.060±2.63df	0.257±0.01	0.9975±0.0032
Sample 16	42.806±3.25f	0.231±0.03	0.9841±0.0069	37.009±2.75gh	0.259±0.02	0.9849±0.0254
Sample 17	31.943±1.26c	0.219±0.06	0.9868±0.0682	27.557±2.45cf	0.261±0.04	0.9989±0.0356
Sample 18	53.522±6.45g	0.198±0.01	0.9876±0.0856	40.364±4.23ij	0.251±0.08	0.9877±0.0568

Sample 19	34.715±2.36ad	0.208±0.11	0.9978±0.0024	29.439±3.25c	0.233±0.02	0.9981±0.0364
Sample 20	43.372±1.25f	0.216±0.06	0.9867±0.0264	38.482±1.30ih	0.241±0.04	0.9892±0.0256
Sample 21	37.450±1.63de	0.245±0.03	0.9923±0.0036	33.041±2.34b	0.272±0.11	0.9926±0.0085
Sample 22	50.378±2.46g	0.219±0.08	0.9848±0.0256	42.204±1.32j	0.254±0.012	0.9881±0.0253
Sample 23	43.264±2.39f	0.225±0.06	0.9846±0.0368	36.876±2.65gh	0.256±0.05	0.9873±0.0785
Sample 24	56.762±2.68	0.182±0.01	0.9651±0.0567	41.977±3.45j	0.245±0.07	0.9881±0.0365

***** *In a column means followed by the same lowercase letters do not differ significantly by the two-way ANOVA and Tukey HSD test $p<0.005$.*

A two-way ANOVA was performed to evaluate the effects of starch and non-starch thickeners on k and n. For k of the upward curve, there was a significant main effect for the starch ($p < 0.001$); no significant main effect for the non-starch thickener ($p > 0.05$) and a significant interaction between starch and non-starch thickeners ($p < 0.001$). For the downward curve k the results were the same: a significant main effect for the starch ($p < 0.001$); no significant main effect for the non-starch thickener ($p > 0.05$) and a significant interaction between starch and non-starch thickeners ($p < 0.001$). For the flow behavior index n factors did not have any significant influences, neither together nor separately. The results from the two-way ANOVA and the Tukey HSD post hoc test are presented in Table 2. Means in a column followed by the same lower-case letters do not differ significantly.

The consistency factor k from the Ostwald-de Waele model can also be used as a viscosity criterion. In terms of this coefficient, all samples thickened with modified potato starch have high viscosity, with the highest being Sample 12 with A(3.8%)+TS(0.2%), which is the most viscous. The lowest value of the coefficient k is the sample with the lowest viscosity Sample 1 with A(3.4%)+GG(0.1%). The results reported for the modified waxy cornstarch samples (Samples 13 to 24) show similar results and the highest k value (highest viscosity) is sample 22 (R(3.8%)+GX(0.2%)). The lowest values were reported for samples 15 and 17 (R(3.4%)+GX(0.1%)/TS(0.1%)). When comparing the values between the ascending and descending curves, it is evident that the samples thickened with A(3.4%)+TS(0.2%), A(3.8%)+TS(0.1%), R(3.4%)+GG(0.2%), R(3.4%) have the greatest changes in combination with TS(0.2%) and R(3.8%)+TS(0.1%). With the smallest deviations, it is evident that they are the samples with Adamil 3.4% with GX 0.1% and 0.2%. The power law equation was an adequate model to describe the flow behavior of the samples in this study.

The flow behavior index, n, informs the deviation of the Newtonian flow for which $n = 1$. This parameter for all samples was below 1 point, indicative of the pseudoplastic (shear-thinning) nature of tomato ketchup (Kumbár et al., 2019). The flow indices (n) of the modified potato starch samples were between 0.158±0.11 (Sample 6 with A(3.4%)+TS(0.2%)) and 0.275±0.06 (Sample 5 with A(3.4%)+TS(0.1%)). For the samples with modified starch from the wax maze, the lowest value of the coefficient is for sample 21 (R(3.4%)+GX(0.1%)), and the highest is Sample 24 (R(3.8%)+TS(0.2%)).

3.2. Bostvik consistency

The Bostwick consistometer is commonly used in ketchup quality control, measuring the flow length (in centimeters) of a product sample in 30 s. The means and standard deviations for Bostwick consistency of the ketchup are presented in Table 3.

Table 3. Bostwick consistency of the ketchup

Ketchup with starch and non-starch thickener, %	**GG (0.1)**	**GG (0.2)**	**GX (0.1)**	**GX (0.2)**	**TS (0.1)**	**TS (0.2)**
A (3.4)	7.27±0.25	5.50±0.00ab*	5.37±0.06bc	4.57±0.15de	6.00±0.20	4.30±0.00f
A (3.8)	6.57±0.06	4.87±0.06g	4.53±0.06de	4.03±0.06h	4.67±0.15d	4.03±0.06h
R (3.4)	5.67±0.12ai	4.40±0.10ef	5.27±0.06cj	5.17±0.06j	5.80±0.26i	3.43±0.06
R (3.8)	5.60±0.10a	3.93±0.06h	4.90±0.10g	2.87±0.06	4.53±0.06de	2.37±0.06

**Means followed by the same lower case letters do not differ significantly by the two way ANOVA and Tukey HSD test ($p<0.05$).A two-factor analysis was performed on all values of starch and thickener to determine the factor influence.*

A two-way ANOVA was performed to evaluate the effects of starch and non-starch thickeners on the ketchup Bostwick consistency. The results indicated a significant main effect for the starch, ($p < 0.001$); a significant main effect for the non-starch thickener ($p < 0.001$) and a significant interaction between starch and non-starch thickener ($p < 0.001$). In Table 3. the means followed by the same lower-case letters do not differ significantly according to the Tukey HSD test.

Since the ketchup model systems contain the same amount of starch, the observed differences (Table 3) may be due to the different botanical origin and/or modification pattern of the starch preparations. In general, corn starch showed lower Bostwick values than potato starch, which is also indicative of the higher viscosity. The same dependence was observed in the analysis of the obtained ketchup samples. The samples with cross-linked esterified distarch adipate from potato starch showed a thinner consistency than the acetylated distarch adipate from waxy maize starch. With the highest value is the sample with A(3.4%)+GG(0.1%) (7.27±0.25 cm) , and with the lowest value R(3.8%)+TS(0.2%) - 2.37±0.06 cm. It is observed that with an increase in the amount of starch, as well as rubber, the viscosity also increases, correspondingly, the Bostwick consistency values decrease. The lowest values were recorded for the combinations with 0.2% TS.

3.3. Syneresis resistance

Potato starches contain more amylose than maize starches, while amylose is absent in waxy maize (Li and Nie, 2016). Retrogradation involves the formation of a gel-like texture by linking amylose chains and forming a double helix and by linking amylopectin chains into double helices. The retrogradation of amylose chains occurs at a much faster rate than that of amylopectin. This was also observed in the analysis of the ketchup samples obtained with different types of modified starch in combination with hydrocolloids. Despite the evidence presented that guar gum and xanthan gum and their mixtures were most successful in reducing the serum release of tomato ketchup (31), in the analysis of the samples obtained, the serum-released liquid was found in ketchup with only modified potato starch (Adamyl), in combination with guar gum, in an amount of 0.1%. After centrifugation of the samples, separation of separation liquid was detected in samples 1 and 7. In sample 1, the liquid was separated in the amount of 17.999±1.023% (on the 5th day); 20.333±1.113% (on the 10th day) and 26.300±0.897% (on the 20th day), and sample 7 -12.881±1.056% (on the 5th day), 13.767±0.876% (on the 10th day) and

14.088±2.345% (on the 20th day). As a comparison, when increasing the amount of GG to 0.2%, no syneresis fluid is released.

4. Conclusions

Based on the conducted analyses, it can be concluded that when comparing the properties of the used modified starches, tomato ketchup with modified waxy corn starch is more stable. In combination with increasing the amount of non-starch thickeners, this stability is enhanced. The best results in terms of rheological properties as well as consistency are observed with a combination of 3.4% modified starch (Resistamil) + 0.2% carrageenan. The international requirements for the Bostwick index are between 7.5 and 10 cm so the concentration of the thickeners should be reduced. This will help to obtain a more resistant product to mechanical impact and storage.

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