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1 **INFLUENCE OF HEALTHY SWEETENERS (TAGATOSE AND**
2 **OLIGOFRACTOSE) ON THE PHYSICOCHEMICAL CHARACTERISTICS OF**
3 **ORANGE MARMALADE**

4 **Susana Rubio-Arreaez¹, Sergio Sahuquillo¹, Juan Vicente Capella², María Dolores**
5 **Ortolá¹, and María Luisa Castelló^{1,3}**

6 ¹Institute of Food Engineering for Development and ²Institute ITACA (ICT
7 Technologies), Universitat Politècnica de València, Camino de Vera, s/n. 46022,
8 Valencia, Spain

9

10 **ABSTRACT**

11 Today's society shows a growing interest in healthy, safe and high nutritional quality
12 food. Thus, in this paper sweet orange marmalades have been developed using healthy
13 sweeteners (tagatose and oligofractose) in different proportions. Analyses of Brix, pH,
14 moisture, water activity, antioxidant capacity, optical and rheological properties have
15 been carried out, initially and after 45 days of storage. Microbiological analyses have also
16 been performed to determine their stability. Furthermore, a sensorial assessment has been
17 conducted to find out acceptance of these new orange marmalades by consumers. The
18 results showed that the highest proportion of oligofractose contributed to improve the
19 initial antioxidant capacity of marmalades. The marmalade with the same proportions of
20 oligofractose and tagatose was more consistent and showed a further increase in the
21 elastic component over time. All marmalades had a similar appearance, but oligofractose
22 reduced L*. Finally, orange marmalades made with healthy sweeteners were better
23 scored.

24

25 **KEYWORDS:** marmalade, tagatose, oligofractose, rheology, color, sensory analysis.

¹Corresponding author: mcasgo@upvnet.upv.es. Universitat Politècnica de València. Camino de Vera s/n. 46022. Valencia. Spain. Phone number: 0034963879967

26 **PRACTICAL APPLICATIONS**

27 The development of new healthier marmalades offer new alternatives to the conventional
28 ones not only to prevent caries and obesity but also to provide functional features
29 associated with the use of tagatose and oligofructose as sweeteners. However, not always
30 it is possible to replace traditional components by others and that is why is so important
31 to assess their technological influence. In this study, the viability of the use of both
32 sweeteners to prepare orange marmalades has been checked.

33

34 **INTRODUCTION**

35 Nowadays, owing to the current lifestyle of society, there is an increasing demand for
36 healthy food products such as fruit and vegetables. However, these products are highly
37 perishable with the consequent problems of distribution and shelf life. As an alternative,
38 processing makes it possible to extend their marketability, whilst maintaining some of the
39 characteristics of the fresh products to a certain extent. In this regard, marmalades are a
40 typical example of more stable fruit-derived products. Among the diverse variety of fruits
41 and the requirements for manufacturing marmalades, oranges may be a good choice in
42 the Mediterranean area as they are readily available at an affordable price, and also
43 because of their high nutritional content.

44 In the case of marmalades, as in many other cases, sugars/sweeteners are crucial for these
45 kind of products to achieve the right texture. In addition, they are responsible for the high
46 concentration of Brix, the reduction of a_w and consequently the control of the microbial
47 growth in the product. Traditionally, sucrose has been used as the main sugar to
48 manufacture marmalades. However, because of the negative connotations associated with
49 sugar consumption (cariogenesis, high caloric intake, increase in the glycemic index,
50 etc...) it has been replaced by bulk sweeteners (polyols) or high-potency sweeteners

51 (saccharine, aspartame...) depending on the properties required in the product (Edwards
52 2002; O'Donnell and Kearsley 2012). Nevertheless, these sweeteners also present some
53 drawbacks. Concretely, most of the polyalcohols have a laxative effect. In the case of
54 high-potency sweeteners, there is a lot of controversy since their relation with the
55 development of different cancers and other diseases is being questioned (Weihrauch and
56 Diehl 2004; Soffritti *et al.* 2006; Renwick and Nordmann 2007).

57 On the other hand, the World Health Organization (2014) considers reducing the
58 excessive consumption of sugars and other carbohydrates of fast absorption, such as
59 sucrose an urgent matter, whilst increasing daily physical activity in order to stop the
60 trend towards obesity and diabetes type 2. A fast absorption of sugar may cause glycemic
61 peaks and the excess sugar may be quickly converted into fat in the organism (Lu *et al.*
62 2008; Lina *et al.* 2002). In fact, most of the sugars (sucrose, fructose and glucose) have
63 around 4 kcal/g, although their glycemic index (GI) **changes**. Thus, glucose has the
64 highest GI, with a value of 100, followed by sucrose with 65 and fructose with a GI of
65 25. Factors such as obesity, diabetes, and the increasing awareness of the need to improve
66 diet, increase the demand for alternative sweeteners to those previously mentioned.
67 Fortunately, the food industry currently offers healthy alternatives, such as tagatose,
68 oligofructose, stevia, isomaltulose, etc... The challenge is to check their viability to
69 reformulate traditional products in order to keep or even improve their technological
70 properties.

71 One of the alternatives to the traditional sweeteners is D-Tagatose (D-tag) that is a
72 ketohexose bulk sweetener, a stereoisomer of D-fructose, with a texture very similar to
73 sucrose, almost as sweet as sucrose, since its sweetening power is 92% (Oh 2007; Taylor
74 *et al.* 2008; Calzada-León *et al.* 2013) but with only 1.5 kcal/g and it does not cause dental
75 caries (Levin 2002). D-tagatose received Generally Recognized as Safe status by the Food

76 and Drug Administration in 2001 and entered the US market as a sweetener in 2003
77 (Donner *et al.* 2010). It is found naturally in several foods, including cheese and yoghurt.
78 It can also be produced from D-galactose by means of a chemical method using calcium
79 as a catalyst (Oh 2007). Tagatose is very suitable for confectionary products, ice creams,
80 soft drinks and breakfast cereals (Vastenavond *et al.* 2011). It is considered a functional
81 food because it is partially metabolized and the part that is not absorbed (80% of the
82 intake) ferments in the colon, where it performs functions as soluble fiber (Taylor *et al.*
83 2008) favouring lactic acid bacteria and *Lactobacillus* specie bacteria (Petersen-Skytte
84 2006).

85 On the other hand, oligofructose is an oligosaccharide derived from sucrose, which acts
86 as dietary fibre regulating intestinal transit. It improves calcium absorption (van den
87 Heuvel *et al.* 1996) and reduces cholesterol and blood sugar levels (Chacón-Villalobos
88 2006). Moreover, it presents a prebiotic effect because it favours the selective growth of
89 lactic bacteria and bifidobacteria (Ledur *et al.* 2013). Oligofructose has approximately
90 between 30% and 60% of the sweetness of sucrose and it is easily hydrolysed by the
91 action of acids or enzymes (Coussement 1999). It is highly soluble and possesses
92 technological properties that are closely related to those of sucrose and glucose syrups
93 (sweet taste, stability...) because it has free sugars (Pimentel *et al.*, 2014). It is often used
94 in combination with high intensity sweeteners. The replacement of carbohydrates by
95 oligofructose offers the advantage of not compromising on taste and texture, while
96 delivering nutritionally enhanced products (Franck 2002).

97 The aim of this paper focuses on characterising sweet orange marmalades formulated
98 with different combinations of tagatose and oligofructose analysing their moisture
99 content, Brix, pH and antioxidant capacity initially and after 45 days of storage. Besides,

100 their rheological and optical properties have been also registered. Finally, a sensorial
101 analysis has been carried out to assess their acceptability by potential consumers.

102

103 **MATERIALS AND METHODS**

104 **Formulations and Manufacturing Processes of Orange Marmalades**

105 Marmalades were produced using 60% orange pulp (*Navelate* variety), 40% sucrose
106 (Azucarera Española, Spain) or healthy sweeteners (tagatose or oligofructose) and 1%
107 agar-agar (Roko Agar^(R), Spain) on the percentage of sucrose or sweeteners. Tagatose was
108 obtained from Damhert Nutrition (Tagatesse ^(R), Belgium) and according to the
109 information of the label it was composed by 39.9% of tagatose, 39.9% of isomalt, 0.02%
110 of sucralose and 20% of dietary fiber (inulin and oligosaccharides). Oligofructose was
111 obtained from Sensus (Frutalose OFP^(R), Netherlands).

112 The following notation was used depending on the combination of sweeteners used:
113 Control marmalade: 100% sucrose, Marmalade A: 50% oligofructose and 50% tagatose,
114 Marmalade B: 30% oligofructose and 70% tagatose, and Marmalade C: 70%
115 oligofructose and 30% tagatose.

116 Oranges collected directly from crop were peeled and mixed with sucrose or the
117 corresponding combination of healthy sweeteners and the agar-agar in a thermal blender
118 (Thermomix, TM31, Vorwerk, Germany). After that, the mixture was cooked at 100 °C
119 for 20 min at 350 rpm. Then, glass jars previously sterilized in an autoclave at 121 °C for
120 15 min, were filled with the marmalade. These jars were turned over to ensure proper
121 sealing for 1 hour. Finally, the marmalade was allowed to cool for 24 hours and in that
122 time jellification took place. Three batches of oranges were used to prepare the
123 marmalades. Triplicated analyses were performed for each batch on the first day of
124 storage and after 45 days of storage.

125

126 **Analytical Determinations**

127 **Moisture Content, Brix, pH and Water Activity.** Moisture content (x^w), was
128 determined gravimetrically by drying approximately 1 g of marmalade to a constant
129 weight in a vacuum oven at 60 °C (method 20.103 AOAC 2000). Water activity (a_w) was
130 determined with a dew point water activity meter Decagon Devices, Inc. (Aqua Lab 4TE,
131 Pullman, Washington, USA). Soluble solid content (Brix) was measured with a
132 refractometer at 20 °C (Atago 3T, Tokyo, Japan) and pH was registered with a pH-meter
133 (Seven Easy, Mettler Toledo, Barcelona, Spain).

134

135 **Antioxidant Capacity.** The antioxidant activity of marmalades was analyzed on the basis
136 of the scavenging activities of the stable 2,2-diphenyl-1-picrylhydrazyl free radical
137 (Brand-Williams *et al.* 1995; Shahidi *et al.* 2006). 1 g of marmalade was mixed with 6
138 mL of pure methanol for 5 min in a vortex, keeping the supernatant. This mixture was
139 centrifuged at 13,000 rpm for 10 min. The absorbance of 3.9 mL of the DPPH solution
140 (0.025 mg/mL, prepared in methanol: water (80:20)) was read at 515 nm in a
141 spectrophotometer Thermo Fisher Scientific, Inc. (Helios Zeta UV-VIS, Waltham,
142 Massachusetts, USA). Then, 0.1 mL of the supernatant was mixed with the methanolic
143 solution of DPPH and absorbance was read again after 30 min. Quantification was
144 performed considering a standard curve of Trolox (6-Hydroxy-2,5,7,8-
145 tetramethylchroman-2-carboxylic acid) and the results were expressed as mg of Trolox
146 equivalent per 100 g of marmalade.

147

148 **Optical Properties**

149 The colour of orange marmalades placed in 20 mm-wide cuvettes was measured using a
150 spectrophotometer Konica Minolta, Inc. (CM-3600d, Tokyo, Japan). CIE-L*a*b*
151 coordinates were obtained using D65 illuminant and 10° observer as reference system.

152

153 **Rheological Analysis**

154 Rheological properties of studied orange marmalades were obtained using a controlled
155 stress rheometer Thermo Fisher Scientific, Inc. (Haake RheoStress 1, Waltham,
156 Massachusetts, USA), at 25 °C. Measurements were carried out with plate–plate geometry
157 and a 2.0 mm gap for steady state and oscillatory tests (Sato and Cunha 2009), by means
158 of steady state essays or oscillatory essays to study the pseudoplastic or viscoelastic
159 behavior of marmalades respectively.

160 Firstly, steady state measurements were performed with a shear rate linearly ranging from
161 0 to 100 s⁻¹, in 3 sweeps (up, down and up-cycles), in order to eliminate thixotropy. The
162 data obtained in the third sweep were fitted to the Herschel–Bulkley model (equation 1)
163 (Peinado *et al.* 2012). This model can describe Newtonian and a large group of time
164 independent non-Newtonian fluids. There are three parameters: τ is the shear stress (Pa),
165 τ_0 is the yield stress above which the fluid starts flowing (Pa), γ is the shear rate (s⁻¹), k is
166 the index of consistency (Pa·s^{*n*}) and n is the index of fluidity (Skelland 1967).

$$167 \quad \tau = \tau_0 + \kappa \cdot \gamma^n \quad (1)$$

168 In second place, an oscillatory assay was carried out following the power-law that
169 described the mechanical spectrum within the linear viscoelastic region in terms of
170 storage (G') and loss (G'') modulus as a function of frequency (equations 2 and 3)
171 (Subramanian *et al.* 2006; Basua *et al.* 2011):

$$172 \quad G' = a \cdot \omega^b \quad (2)$$

$$173 \quad G'' = c \cdot \omega^d \quad (3)$$

174 Where, ω is the angular speed ($\text{rad}\cdot\text{s}^{-1}$), a is the low frequency storage modulus (Pa^b);
175 b is the power-law index for the storage modulus (dimensionless); c is the low frequency
176 loss modulus (Pa^d); and, d is the power-law index for the loss modulus (dimensionless).
177 The value of the shear stress to fulfill the linearity of G' and G'' was obtained in a
178 preliminary trial. In order to do so, an interval of shear $\tau = 0.1\text{-}10$ Pa was studied and
179 three fixed frequencies were marked in the range of 0.1-10 Hz. Once these 3 curves were
180 represented, the lineal zone of viscosity was obtained. Having chosen the value of τ in
181 the linear zone for all the frequencies, the oscillatory assay was performed between 0.1-
182 10 Hz (Peinado *et al.* 2012).

183

184 **Microbiological Analysis**

185 Serial dilutions were prepared by homogenising 10 g of marmalade with 90 mL of 1%
186 sterile peptone water in a stomacher bag, using sterile techniques. Mesophilic aerobic
187 populations were analysed in Plate Count Agar (Scharlau Chemie, 1-329, Barcelona,
188 Spain) incubating samples for 72 hours at 31 °C. Yeast and moulds were determined in
189 Sabouraud Chloramphenicol Agar (Scharlau Chemie, 1-166, Barcelona, Spain) plates for
190 5 days at 31 °C. Samples were analysed at 45 days of storage.

191

192 **Sensorial Analysis**

193 An acceptance test using a 9-point hedonic scale (ISO 4121:2003) was used to evaluate
194 the following attributes: colour, aroma, texture, consistency, spreadable capacity,
195 palatability, flavor, sweetness, bitterness, and global preference (ISO 5492:2008) in the
196 three formulations with different combinations of healthy sugars (A, B and C) along with
197 the control marmalade. The possible appearance of sineresis was also assessed. Moreover,
198 the intention of buying was considered. The panel consisted of 30 trained panelists, in the

199 age range of 20-50 years old, who are regular consumers of this kind of marmalades.
200 Samples were presented in jars of 25 mL presented one after another. Three testing
201 sessions were conducted in a sensory evaluation laboratory built according to the
202 international standards for test rooms (ISO 8589: 2007).

203

204 **Statistical Analysis**

205 Statgraphics plus (version 5.1) software was used to perform the statistical analyses.
206 Analyses of variance (multifactor ANOVA) were carried out to discern whether the effect
207 of the formulation or the time of storage on the studied marmalades was significant. The
208 interactions between factors were considered.

209

210 **RESULTS AND DISCUSSION**

211 **Compositional Characterization of Marmalades**

212 Table 1 shows the results of moisture content (x^w), Brix, water activity (a_w), pH and
213 antioxidant capacity. In all cases the sugar concentration reached in marmalades was 50
214 Brix and it remained during the storage period considered. Moreover, marmalades B and
215 C showed higher water content than the control and marmalade A. With time, the
216 moisture content of samples B and C reduced, while in marmalade A it increased though
217 in all cases there were slight differences. Regarding water activity, storage time was the
218 factor which implied the most significant influence, showing a small decrease in water
219 activity at 45 days of storage, except for sample A. Focusing on the pH, all marmalades
220 showed values below 3.8, which would ensure proper microbiological stability. For all
221 analyzed products, pH ranged between 3 and 4, in the same magnitude of order as the pH
222 of other jams made using strawberry, peach, plum and apricot (Carbonell *et al.* 1991;
223 García-Martínez *et al.* 2002). Furthermore, in the control marmalade and sample A pH

224 decreased after storage as was also observed by Rababah *et al.* (2011) in strawberry jams
225 with sucrose. However in the same paper the pH value of orange fruit after processing to
226 obtain jam was decreased significantly, and only after 5 months of storage there was again
227 a significant decrease in pH. In terms of antioxidant capacity, initially formulation C
228 showed the highest value probably due to the greater amount of oligofructose in its
229 composition, in contrast with the results of Ścibisz and Mitek (2009) who observed that
230 high bush blueberry jams had the lowest levels of anthocyanins and total phenolics when
231 oligofructose was included in their formulation. In our research, both formulation A and
232 B reported lower antioxidant content than the control, showing that tagatose reacts less
233 than table sugar with free radicals. These results differ from those reported by Zeng *et al.*
234 (2012) who observed that via the Maillard reaction, rare sugars (especially D-tagatose)
235 induced a more remarkable improvement than D-fructose in the radical scavenging
236 activity and oxidation–reduction potential of the hydrolysates of tune backbone.
237 Moreover, in our study, the antioxidant capacity of marmalades increased over time,
238 showing possible combinations of components which would imply the appearance of new
239 antioxidants. However, Rababah *et al.* (2011) observed that antioxidant activity of orange
240 marmalades prepared with sucrose decreased significantly after 3 and 4 months, and 5
241 months, respectively.

242

243 **Rheological Properties**

244 The rheological properties of the studied marmalades were determined by two tests:
245 steady and oscillatory obtaining the parameters of the models considered in each case.
246 The results obtained in the stationary test are presented in Figure 1. Rheograms indicate
247 that initially there were no differences between samples, except for formulation B which
248 scored slightly higher than the rest. After the storage period, the shear stress exceeded

249 was reduced in all cases although to a lesser extent in marmalade A. Furthermore, in Table
250 2 the parameters of the Herschel-Bulkley model are shown. As expected, all marmalades
251 showed a shear thinning behavior ($n < 1$). However, the yield stress (τ_0) was similar in
252 marmalade control and formulation A initially and after 45 days of storage whereas in
253 formulations B and C the values of this parameter were significantly lower especially
254 after storage in B. Besides, marmalade A showed the highest level of consistency after
255 storage, giving evidences that the combination of oligofructose and tagatose in same
256 proportions would improve consistency of marmalades during the storage. With respect
257 to the index of fluidity (n), no significant differences were found considering the
258 formulation studied but the time implied a reduction of this index in all cases. In studies
259 carried out by other authors (Peinado *et al.* 2012; Rosa *et al.* 2009) where sugars were
260 also replaced by other sweeteners (in this case isomaltulose) a decrease in consistency
261 and cohesiveness of strawberry jams was observed **respect to the sucrose-jams**. In our
262 study, only an increase of consistency was observed in the combination of 50%
263 oligofructose and tagatose, but not for the other blends.

264 Figure 2 shows the rheological results of the oscillatory assay where the frequency
265 dependence on storage (G') and loss (G'') moduli of the orange marmalades formulated
266 with healthy sweeteners are represented. This type of test determines the ratio between
267 the elastic and viscous component of a material and it is useful to quantify to what extent
268 it behaves as a solid or liquid. Since in all cases G' was greater than G'' , marmalades
269 showed a semi-solid behavior. This is a typical gel characteristic being more elastic than
270 viscous (Peinado *et al.* 2012).

271 Concerning the formulations studied, at the beginning of storage there were no differences
272 between the control marmalade and the marmalade with the same amount of tagatose and
273 oligofructose (formulation A), for both moduli (G' and G''). Moreover, marmalades B

274 and C were similar in terms of viscous level, whereas the most elastic character was for
275 marmalade with more oligofructose (C). At the end of storage, the increase in the elastic
276 component of formulation A was noteworthy, unlike what was observed in the other
277 marmalades, giving evidence of the interaction between the analyzed factors as a function
278 of the sweeteners used. Besides in this assay, marmalade B had more similar elastic
279 characteristics to the control marmalade.

280 In order to quantify in depth the differences between the oscillatory test of the analyzed
281 samples, the values of both the storage (G') and the loss (G'') moduli were fitted with
282 respect to the angular speed (ω) with the power-law model as described in materials and
283 methods. The parameters of this model are shown in Table 2. As can be observed, initially
284 there were no significant differences between samples in a and b parameters of the power-
285 law model for the storage modulus. However, in marmalade A a increased significantly
286 after 45 days of storage, while the time factor did not affect the other formulations. This
287 behavior would be consistent with that observed in Fig. 2, reflecting a more elastic nature
288 of marmalade A at the end of storage. Regarding the terms related to the loss modulus (c
289 and d) it should also be mentioned that marmalade A had a significantly greater value of
290 c in coherence with the position of its curve G'' versus frequency (ω) above the rest of
291 the samples (Fig. 2). However, these differences were much lower than in the case of the
292 parameters related to the storage modulus. In addition, parameter d fluctuated more
293 between the cases studied. In fact lower values were only observed in the formulation A
294 with regard to the initial control and at the end of storage and higher values in marmalade
295 B after 45 days. In contrast to the few differences found in this study, Peinado *et al.* (2012)
296 observed that by replacing sucrose for isomaltulose in the formulation of different
297 strawberry spreadable products resulted in a decrease in parameters a and c of the power-
298 law model. This decrease was associated with how sugar type influences the availability

299 of water in the mixture of pectin-sugar-acid and therefore in the formation of hydrogen
300 bonds and the possible association of water in the pectin polymer chain. In this study, the
301 gelling agent used was agar-agar instead of pectin and it could have homogenized the
302 rheological properties of the marmalade regardless of the type of sugar used, except in
303 the case of marmalade A.

304

305 **Optical Properties**

306 Colorimetric coordinates b^* and a^* of the different studied sweet orange marmalades
307 initially and at the end of storage placed in the chromatic plane are shown in Figure 3A.
308 The new marmalades, especially for A and B formulations, increased b^* and a^*
309 coordinates in comparison with the values of the control marmalade. Consequently the
310 chrome ($C^*=(a^{*2}+b^{*2})^{1/2}$) followed the same trend, while the hue ($h^*=\arctg(b^*/a^*)$)
311 remained very similar to the control giving place to a similar appearance in all cases.
312 Furthermore, values of initial and final luminosity (L^*) of the studied marmalades are
313 also presented. According to these results, initially, L^* of samples A and especially B was
314 much higher than in the control marmalade unlike what happened to marmalade C.
315 Therefore, high concentrations of oligofructose significantly reduced L^* of marmalades.
316 Considering the time factor in L^* , the most stable formulations were A and C, while L^*
317 in the control and B changed oppositely, increasing in the case of control and decreasing
318 for B.

319 In other studies, Peinado *et al.* 2015 showed that strawberry jams formulated with the
320 healthier sugar isomaltulose and different concentrations of citric acid and pectin,
321 darkened with time. Besides that, the colorimetric coordinates of the products containing
322 the sucrose-isomaltulose blend seemed to be influenced by the percentages of pectin and
323 citric acid while the colour of the samples containing the fructose-isomaltulose blend, did

324 not seem to be affected by the different variables. Therefore the influence of the different
325 ingredients on the food system does not only depend on their concentration or distribution
326 within the different system phases but also on the different component interactions during
327 the studied period (Dervisi *et al.* 2001; Renard *et al.* 2006; Peinado *et al.* 2015).

328

329 **Microbiological Analysis**

330 There were no colonies of molds and yeast or aerobic mesophilic found in any of the
331 marmalades in this study during the storage period considered.

332

333 **Sensory Analysis**

334 Figure 4 shows a radial chart of the average scores for each attribute evaluated (color,
335 aroma, texture, consistency, spreadable capacity, palatability, flavor, sweetness and
336 bitterness) besides the global preference and intention of buying of the studied
337 marmalades. As can be seen, A and B formulations showed the highest scores in all
338 attributes, although no significant differences were found in color, aroma, texture and
339 consistency among the samples. Furthermore, A and B, which had higher proportions of
340 tagatose, showed the highest sweetness. This would be consistent with the
341 recommendations given by the manufacturer of the commercial tagatose (two tablespoons
342 of sucrose provides the same sweetness as one tablespoon of tagatose), although as was
343 mentioned in the introduction, tagatose should have similar sweetening power to sucrose
344 (Oh 2007; Taylor *et al.* 2008; Calzada-León *et al.* 2013). However, the commercial
345 tagatose used in this study was composed also by oligosaccharides, isomalt and sucralose.
346 The higher sweetening power of this combination could be due to the synergic influence
347 between these sweeteners. In fact, according to the Patent EP0946112 B1 (Dörr and Jager
348 2002), oligosaccharides increase the sweetness and improve the taste of an acesulfame-

349 k/aspartame mixture. Moreover, although the level of bitterness in the marmalades
350 evaluated in this study was expected to be very low, analyzing the possible interference
351 of the combination of sweeteners used in this property was deemed relevant. Thus, the
352 control and formulation C had the highest bitterness, showing the great ability of tagatose
353 to hide this taste. Furthermore, no marmalade developed syneresis. In relation to the
354 rheological properties, the results obtained in the sensorial analysis were in accordance
355 with those registered instrumentally. Finally, attention should be brought to the fact that
356 both the global preference and buying intention of all marmalades formulated with
357 healthy sweeteners were higher than those containing only sucrose.

358

359 **CONCLUSIONS**

360 The reformulation of orange marmalade with healthy sweeteners such as tagatose and
361 oligofructose is feasible. Only oligofructose improved the antioxidant capacity compared
362 to marmalade prepared with sucrose and also reduced its luminosity. In general, all
363 marmalades had the same appearance. In terms of rheology, formulation with the same
364 proportion of tagatose and oligofructose improved consistency and elastic component of
365 marmalades over time. Moreover, all of them reported microbiological stability in storage
366 during the storage period considered. Finally, global acceptance and intention of buying
367 of marmalades with healthy sweeteners were higher than for marmalade containing only
368 sucrose.

369

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373

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483

484

485 **FIGURE CAPTIONS**

486

487 **FIG. 1.** MEAN FLOW CURVES (RHEOGRAMS) OBTAINED FROM THE STEADY
488 ASSAY OF ORANGE MARMALADES AT THE BEGINNING AND AT THE END
489 OF STORAGE. SAMPLES WERE CODED RESPECT TO THE AMOUNT OF
490 SUGARS AS: CONTROL (100% SUCROSE); A (50% OLIGOFRUCTOSE AND 50%
491 TAGATOSE); B (30% OLIGOFRUCTOSE AND 70% TAGATOSE), AND C (70%
492 OLIGOFRUCTOSE AND 30% TAGATOSE).

493

494 **FIG. 2.** AVERAGE FREQUENCY CURVES OBTAINED IN THE OSCILLATORY
495 TEST OF ORANGE MARMALADES AT THE BEGINNING AND END OF
496 STORAGE. SAMPLES WERE CODED RESPECT TO THE AMOUNT OF SUGARS

497 AS: CONTROL (100% SUCROSE); A (50% OLIGOFRUCTOSE AND 50%
498 TAGATOSE); B (30% OLIGOFRUCTOSE AND 70% TAGATOSE), AND C (70%
499 OLIGOFRUCTOSE AND 30% TAGATOSE). EMPTY SYMBOLS REFER TO
500 VALUES OF STORAGE MODULUS (G') AND FILLED SYMBOLS REFER TO
501 VALUES OF LOSS MODULUS (G'').

502

503 **FIG. 3.** A) CHROMATIC PLANE REPRESENTATION (b^*-a^*) OF THE STUDIED
504 MARMALADES INITIALLY AND AFTER 45 DAYS OF STORAGE. Straight line
505 represents the hue of control orange marmalade for day 1 from (0,0). B) LUMINOSITY
506 (L^*) OF THE DIFFERENT FORMULATIONS OF MARMALADE INITIALLY AND
507 AFTER 45 DAYS OF STORAGE. SAMPLES WERE CODED RESPECT TO THE
508 AMOUNT OF SUGARS AS: CONTROL (100% SUCROSE); A (50%
509 OLIGOFRUCTOSE AND 50% TAGATOSE); B (30% OLIGOFRUCTOSE AND 70%
510 TAGATOSE), AND C (70% OLIGOFRUCTOSE AND 30% TAGATOSE). Equal letters
511 indicate homogeneous groups.

512

513 **FIG. 4.** RESULTS OF THE SENSORY ANALYSIS IN THE EVALUATION OF THE
514 SAMPLES CODED RESPECT TO THE AMOUNT OF SUGARS AS: CONTROL
515 (100% SUCROSE); A (50% OLIGOFRUCTOSE AND 50% TAGATOSE); B (30%
516 OLIGOFRUCTOSE AND 70% TAGATOSE), AND C (70% OLIGOFRUCTOSE AND
517 30% TAGATOSE). * p-value <0.05, ** p-value <0.01

518

519 **TABLE 1.** VALUES FOR MOISTURE CONTENT (x^w), Brix, WATER ACTIVITY
520 (a_w), pH AND ANTIOXIDANT CAPACITY OF ORANGE MARMALADES
521 INITIALLY AND AFTER 45 DAYS OF STORAGE.

522

523 **TABLE 2.** RHEOLOGICAL PARAMETERS OF THE HERSCHEL-BULKLEY
524 MODEL AND PARAMETERS OF THE POWER-LAW MODEL FOR
525 MARMALADES INITIALLY AND AT THE END OF STORAGE.