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Gonzalez-Zamora, F.; Igual Ramo, M.; Camacho Vidal, MM.; Martínez Navarrete, N. (2018). Impact of Temperature, Gum Arabic and Carboxymethyl Cellulose on Some Physical Properties of Spray-Dried Grapefruit. International Journal of Food Engineering. 14(5-6). https://doi.org/10.1515/ijfe-2017-0387



The final publication is available at http://doi.org/ 10.1515/ijfe-2017-0387

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Additional Information

# IMPACT OF TEMPERATURE, GUM ARABIC AND CARBOXYMETHYL CELLULOSE ON SOME PHYSICAL PROPERTIES OF SPRAY-DRIED GRAPEFRUIT

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#### **Abstract**

Spray-dried fruit powder may be an interesting alternative for the purposes of promoting fruit consumption among consumers. The use of carrier agents is especially necessary for the production of spray-dried fruit powders. As they may affect some physical properties of the powder, it is important to adjust the amount at which they have to be added to the minimum in order to achieve the necessary effects. The final aim of the study was to identify the most suitable atomization temperature, as well as the optimal concentration of gum Arabic (GA) and carboxymethyl cellulose (CMC) to be used as carriers, in order to obtain grapefruit powder with the maximum dry matter yield (DMY) and porosity, the minimum water content and, simultaneously, with suitable color characteristics. The results of the study don't recommend the use of CMC and suggest that the best color, the one that corresponds to a free-flowing powder, corresponds to a very luminous one, low in chroma and with a hue that is much more yellow than reddish orange.

**Keywords**: grapefruit powder, porosity, yield, luminosity, chroma, hue angle.

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#### 1. Introduction

In recent years, the health benefits of eating fruit and vegetables have been linked to their role in the preven- tion of different diseases. These protective effects are attributed to the presence of bioactive compounds with antioxidant capacity that affects the reduction of degenerative diseases [1]. However, the main drawback to the consumption of fresh fruit and vegetables is their short lifespan. In the mature state, they present a high water content, making them more susceptible to decomposition by microorganisms, chemical and enzymatic reactions [2]. In fact post-harvest losses of well over 50 % may be generated while marketed [3]. In this sense, one challenge faced by the food industry is that of researching and developing processes and/or products that, in addition to being safe, maintain the health benefits of fruit at a maximum, considering the current consumer demand.

Dehydration processes have been widely used in the food industry to obtain products with a greater stability and lower volume, which are easier to handle [4]. Nevertheless, conventional processes should be optimized or new processes should be developed to ensure the best quality of the obtained products. Fruit powder may be an interesting alternative for the purposes of promoting fruit consumption among consumers, either as an ingredient in other foods or after being rehydrated to obtain a juice. Spray-drying is a simultaneous heat and mass transfer operation that implies the change of a food from a liquid state to a dry particulate state.

Despite the obtained powder will benefit from great biochemical and microbiological stability, it could present some problems as regards its physical stability. Spray drying is a rapid dehydration method that frequently leads to the obtaining of an amorphous matrix, glassy or rubbery depending on the glass transition temperature (Tg). Powdered foods in the rubbery state may exhibit stickiness and caking problems [5]. This structural collapse is characterized by a sharp loss in porosity which affects aroma retention or rehydration capacity, among other things [6]. The rubbery state is especially frequent in powdered fruit products, related to their composition. Most fruit soluble solutes are low molecular weight sugars and organic acids all of which have a low Tg [7]. This leads to the Tg of the product being easily exceeded during fruit spray-drying, and also during storage, with the consequent adhesion of rubbery powder particles both to each other and to the equipment, decreasing the product yield and causing operational problems [8]. In general, the stickiness causes considerable economic loss and limits the application of spray drying on foods as well as on pharmaceutical materials [9].

Some of the problems associated with the rubbery state can be solved by adding biopolymers that act as process carriers and confer stability on the product. High molecular weight carbohydrates, such as starches, maltodextrins and gums, many of them capable of increasing the Tg, are included among these carriers [10, 11]. Although these are the materials most commonly used for this purpose, it is also true that large amounts of carbohydrates are required to avoid fruit powder stickiness, which increases the cost and may alter the original flavor, taste and color of the product [12]. According to these authors, in addition, the films that they form are very easily moisturized which, while necessary to ensure a good rehydration of the powder product, makes it less advisable to use them for the purposes of ensuring their stability. For this reason, the search for other types of drying aids is still ongoing. In this sense, the use of a small amount of proteins, which are amphiphilic in character, has been described [12–16]. An interesting role for carboxymethyl cellulose (CMC), an organic derivative of cellulose, has also been described, involving sugar crystallization. This could reduce the phenomenon of stickiness, considering the fact that crystalline sugar has a lower water sorption potential, despite the fact that solubility decreases [2]. Among the more widely used high molecular weight carbohydrates, the natural plant exudates of Acacia trees, the gum Arabic (GA) is the only gum used in food products that exhibits high solubility and low viscosity in aqueous solution, making the spray drying process easier [17]. GA is especially effective because of its emulsifying properties, due to its low protein content [18]. In this sense, it may be interesting to combine GA with CMC for fruit powder production.

Despite the use of carrier agents being especially necessary for the purposes of producing spray dried fruit powders, they can also affect some physical properties of the powder, such as the water content, porosity or color. For this reason, it is of great interest to select the final use of the powder in order to adjust the quantity of the carriers to be added as much as possible. Whatever the final use may be, a maximum process yield is desirable. Nevertheless, if a tablet is going to be produced, the color or flow properties are less important than if the powder is going to be offered to the consumer to be rehydrated. In the latter case, the yield should be maximized by affecting the desirable physical properties of the powder as little as possible.

In the spray-drying operation a balance between the mass of inlet feed and the mass of powder recovered, taking into account the water eliminated and the powder lost mainly due to the sticking onto the dryer chamber wall is established. In this sense, the process yield or the drying yield can be considered. As for the process yield, to refer the grams of

the solids in the total recovered powdered product to the grams of feed solid-content is recommendable [8, 14, 18–22].

As regards the physical properties, the water content of the obtained powder should be as low as possible. Porosity plays an important role in the agglomerate strength of dried foods [23]. As far as its evolution is concerned, a greater porosity or, what is the same, a lower apparent density, corresponds both to a more free- flowing powder with a greater air volume distributed among particles and also to a more soluble one [18, 23]. On the other hand, it is not easy to know in advance the most desirable expected color in the fruit powder. In many cases, it is not possible to obtain a spray-dried fruit with a color similar to that of the fresh fruit because of the aforementioned need to add carriers in order to avoid operational problems. When no carriers, or a small amount of them, are added, a paste like structure is obtained instead of a powder. In this sense, to look for the natural color in the powder may not be adequate. On the other hand, the carriers lead to color changes in the product [13, 24]. The problem is whether the luminosity, hue and chroma color attributes should take higher or lower values than those of fresh fruit when carriers have been added.

In this sense, the aim of this study was to identify the most suitable atomization temperature, as well as the optimal concentration of GA and CMC to be used as carriers, in order to obtain grapefruit powder with the maximum dry matter yield (DYM) and porosity, the minimum water content and, simultaneously, suitable color characteristics. To this end, it is necessary first to establish whether atomized grapefruit powder of the highest quality relates to the maximum or minimum value of the color attributes.

#### 2. Materials and methods

#### 2.1. Raw material

This study was carried out with grapefruit (*Citrus paradise* var. Star Ruby). GA (Scharlau, Spain) and CMC (Alfa Aesar, Germany) were added to the liquidized grapefruit.

# 2.2. Preparation of feed mixture and spray drying conditions

Grapefruit was washed, peeled (carefully removing the albedo) and liquidized. Liquidized grapefruits were mixed with a water solution containing GA and/or CMC. Solutes were added to water according to the generated experimental design obtained from the response

surface methodology (RSM, Table 1) and commented on below. Liquidized grapefruit (500 g) was mixed with 500 g of each one of these solutions. The mixture was stirred for 30 min until homogeneity was reached. The samples were immediately frozen at -40 °C until atomization. Thawed samples (for 24 h at 8 °C) were fed into a Büchi B-290 (Switzerland) mini spray dryer with the following operating conditions: aspirator of 35 m<sup>3</sup>/h; air flow rate 473 L/h with a co-current flow; pump flow rate 9 mL/min. Drying inlet air temperature was varied according to experimental design (Table 1). After the completion of the experiment and when the air inlet temperature fell below 50 °C, the samples were collected from the product collection vessel.

**Table 1:** Matrix of the central composite experimental design applied and the experimental results ( $\pm$  SD) of water content ( $x_w$ ), porosity ( $\epsilon$ ), dry matter yield (DMY), lightness ( $L^*$ ), hue angle ( $h^*_{ab}$ ) and chroma ( $C^*_{ab}$ ) of the spray-dried powders.

T(1)	GA(2	CMC <sup>(3)</sup>	X <sub>w</sub> <sup>(4)</sup>	٤ (%)	DMY <sup>(5)</sup>	L*	h <sub>ab</sub>	C* ab
150	8.0	1.0	0.029 ± 0.003	71.6 ± 0.2	32.48	90.5 ± 0.2	82.6 ± 0.4	$9.7 \pm 0.2$
150	8.0	1.0	$0.027 \pm 0.003$	72.0 ±	34.86	89.9 ±	78.0 ±	$9.7 \pm 0.3$
100	0.0	4.0	0.027 2 0.000	0.3	54.00	0.2	0.5	J.17 - 4 - 011
150	8.0	1.0	$0.033 \pm 0.006$	74.7 ±	34.45	90.8 ±	88.3 ±	$9.5 \pm 0.3$
			01100 2 01100	0.2		1.7	0.4	
150	8.0	1.0	$0.063 \pm 0.003$	72.6 ±	35.57	91.5 ±	83.5 ±	10.3 ±
				0.2		0.2	0.2	0.2
150	8.0	1.0	$0.034 \pm 0.010$	73.8 ±	41.96	91.4 ±	83.8 ±	10.1 ±
0.000		10.10		0.4		0.2	0.7	0.2
150	8.0	1.0	$0.073 \pm 0.002$	74.1 ±	37.50	91.4 ±	83.9 ±	11.2 ±
33.0				0.2		0.2	0.4	0.2
150	8.0	1.0	$0.079 \pm 0.013$	74.7 ±	31.34	90.91 ±	83.8 ±	11.8 ±
				0.6		0.5	0.4	0.3
150	8.0	1.0	$0.076 \pm 0.010$	74.6 ±	36.20	91.5 ±	85.9 ±	10.5 ±
				0.5		0.3	0.5	0.2
150	8.0	1.0	$0.067 \pm 0.002$	72.4 ±	33.09	89.9 ±	77.8 ±	10.9 ±
				0.3		0.4	0.3	0.2
150	14.7	1.0	$0.142 \pm 0.004$	68.6 ±	37.67	91.2 ±	86.3 ±	$8.6 \pm 0.3$
				0.2		0.3	0.2	
120	12.0	0.0	$0.096 \pm 0.002$	$70.16 \pm$	55.12	90.3 ±	77.4 ±	10.3 ±
				0.6		0.2	0.3	0.3
180	12.0	2.0	$0.057 \pm 0.002$	68.8 ±	25.40	89.9 ±	78.5 ±	$11.8 \pm$
				0.4		0.2	0.3	0.2
150	8.0	2.7	$0.123 \pm 0.007$	75.2 ±	16.88	88.6 ±	72.2 ±	12.3 ±
				0.9		0.3	0.2	0.3
200	8.0	1.0	$0.036 \pm 0.002$	76.2 ±	28.44	88.0 ±	76.9 ±	13.8 ±
				0.8		0.2	0.3	0.2
180	4.0	0.0	$0.079 \pm 0.003$	45.3 ±	35.90	66.5 ±	63.9 ±	$30.4 \pm$
				1.5		1.9	0.6	0.3
100	8.0	1.0	$0.181 \pm 0.002$	68.2 ±	26.20	86.2 ±	79.9 ±	$10.7 \pm$
				0.3		0.2	0.2	0.2
120	4.0	0.0	$0.110 \pm 0.003$	$64.6 \pm$	41.28	82.3 ±	$68.5 \pm$	20.9 ±
				0.4		0.5	0.2	0.4
120 1	12.0	2.0	$0.178 \pm 0.002$	70.6 ±	22.34	90.1 ±	$78.4 \pm$	$9.5 \pm 0.2$
				0.6		0.2	0.7	
180	12.0	0.0	$0.204 \pm 0.012$	69.5 ±	60.94	88.7 ±	74.8 ±	11.4 ±
				0.3		0.3	0.2	0.2
180	4.0	2.0	$0.045 \pm 0.003$	78.2 ±	21.58	87.2 ±	$77.9 \pm 0$ .	17.8 ±
				0.4		0.2	3	0.3
150	8.0	0.0	$0.116 \pm 0.004$	$76.0 \pm$	63.98	90.2 ±	79.1 ±	$11.6 \pm$
				0.4		0.2	0.2	0.2
120	4.0	2.0	$0.115 \pm 0.002$	$76.5 \pm$	17.60	$84.7 \pm$	66.6 ±	14.5 $\pm$
				0.4		0.9	0.2	0.2

150	1.3	1.0	$0.074 \pm 0.003$	64.4 ±	25.28	80.3 ±	63.5 ±	21.7 ±
				1.0		0.7	0.4	0.5

<sup>(1)</sup> Inlet temperature (°C), (2) Gum Arabic (g/100g<sub>liquefled grapefrust</sub>), (3) Carboxymethyl cellulose (g/100g<sub>liquefled grapefrust</sub>),

# 2.3. Experimental Design

For this study, a central composite design (CCD) and the RSM were applied to evaluate the effect of three process independent variables on six response variables, mainly related to the profitability of the process and the quality of the powder, and to propose which levels of the former are optimum for the purposes of achieving the best powder [25]. As independent variables, the inlet air temperature (T,  $100-200\,^{\circ}$ C) and the concentration of gum Arabic (4–12 g GA/100 g liquidized grapefruit) and carboxymethyl cellulose (0–2 g CMC/100 g liquidized grapefruit) were selected. The response variables considered were those of water content ( $x_W$ ), porosity ( $\varepsilon$ ), luminosity ( $L^*$ ), chromatic  $a^*$  and  $b^*$  color coordinates, DMY. Twenty-three experimental runs were generated based on the corresponding rotable and orthogonal CCD (Table 1). The experiments were randomized.

## 2.4. Analysis of response variables

The powder's water content was determined, in triplicate, using the gravimetric method in a vacuum oven (VACIOTEM, JP Selecta, Spain) at  $60\,^{\circ}$ C, p <  $100\,^{\circ}$ mm Hg until constant weight. As the GA and CMC content of each sample was different (Table 1), the water content of the powders was referred to the grapefruit's own solutes (GS) (eq. (1) and (2)) to make the results comparable.

$$x_{w} = \frac{x_{w}^{p}}{(1 - x_{w}^{p})(x_{GS/TS})}$$
 (1)

$$x_{GS/TS} = \frac{m_L (1 - x_w^L)}{m_{GA} + m_{CMC} \pm m_L (1 - x_w^L)}$$
 (2)

where:  $x_w$  is the water content of the powder referred to grapefruit's own solutes (GS, w/w),  $x_w^P$  is the water content of powder (w/w),  $x_{GS/TS}$  is the mass fraction of GS to total

<sup>(4)</sup> gwater/100ggrapefruit's own solutes (5) g powder dry matter/100 g feed dry matter.

sample solutes,  $m_{GA}$ ,  $m_{CMC}$  and  $m_L$  are the mass of GA, CMC and liquidized grapefruit, respectively, in the sample and  $x_w^L$  is the water content of the liquidized grapefruit (w/w).

The porosity, or percentage of air volume related to total volume, was calculated from the true and bulk densities (eq. (3)). The true density ( $\rho$ ) of the product was calculated from its individual components. In this case, water and carbohydrates, both the grapefruit's own and those added, were considered to be the main components of the samples (eq. (4)). For the purposes of bulk density ( $\rho_b$ ) determination, in triplicate, approximately 2 g of the powder were transferred to a 10mL graduated test tube and stirred for 10 s at 1600 rpm in a Vortex (Velp WX F202A0230, Italy). The bulk density was calculated as the ratio mass of the powder to the occupied volume in the tube after stirring.

$$\varepsilon = \frac{\rho - \rho_b}{\rho} \tag{3}$$

$$\frac{1}{\rho} = \frac{x_w^p}{\rho_w} + \frac{x_{CH}^p}{\rho_{CH}} \tag{4}$$

where  $\epsilon$  is the porosity;  $\rho$  and  $\rho_b$  are the true and bulk densities, respectively;  $x_i^p$  and  $\rho_i$  are the mass fraction and density, respectively, of water (i=w) and carbohydrates (i=CH) of the powder, with  $\rho_w$  (20°C) = 0.9976 g/cc and  $\rho_{HC}$  (20 °C) = 1.4246 g/cc [26].

The CIE L\*a\*b\* color coordinates, hue angle (h\*ab, eq. (5)) and chroma (C\*ab, eq. (6)) of the samples were measured in triplicate using a spectrocolorimeter (MINOLTA, CM3600-D, Spain) with a reference illuminant D65 and 10° observer.

$$h_{ab}^* = \operatorname{arctg} \frac{b^*}{a^*} \tag{5}$$

$$C_{ab}^* = \sqrt{a^{*2} + b^{*2}} \tag{6}$$

The percentage of DMY, was determined using eq. (7).

$$DMY = 100 \frac{m^{p} * (I - x_{w}^{p})}{m^{*} (I - x_{w})}$$
 (7)

where  $m^p$  and  $x_w^p$  are the mass (g) and the water content (w/w), respectively, of the obtained powder; m and  $x_w$  are the mass (g) and the water content (w/w) of the product coming into the spry-dryer.

#### 3. Results and Discussion

The water content of the liquidized grapefruit used for the study was  $0.912 \pm 0.009$  g/g. Table 1 shows the experimental results of the different response variables measured in the powders. A second order quadratic equation was used to express the response variables as a function of the independent ones (eq. (8)). Only the significant model terms (p < 0.05) were considered in the final reduced model. Table 2 shows the regression coefficients of the models with a significant correlation of the response variables with the independent ones. The determination coefficients, all in the range of 58–92, indicate that over 50 % of the response variation may be explained in terms of all three independent variables.

$$Y_{i} = \beta_{0} + \beta_{1}X_{1} + \beta_{2}X_{2} + \beta_{3}X_{3} + \beta_{11}X^{2}_{1} + \beta_{22}X^{2}_{2} + \beta_{33}X^{2}_{3} + \beta_{12}X_{1}X_{2} + \beta_{13}X_{1}X_{3} + \beta_{23}X_{2}X_{3}$$
(8)

**Table 2:** Regression coefficients, adjusted determination coefficient  $(R^2)$  and standard error of the estimate (EE) for the polynomial model fitted to predict the response variables as a function of the independent variables.

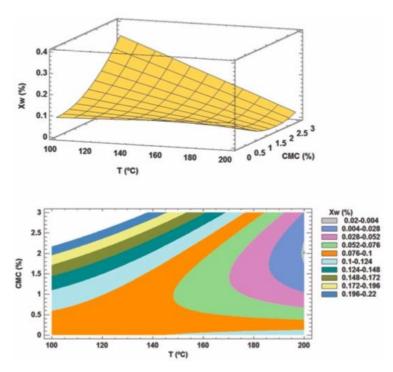
	$\mathbf{x}_{\mathbf{w}}$	ε	DMY	L*	h <sub>ab*</sub>	$C_{ab}^*$
$\beta_0$	0.021	45.38	-39.83	36.38	6.34	53.33
$\beta_1$	0.0002		1.01	0.45	0.66	-0.28
$\beta_2$	0.006	4.46	2.20	3.81	4.50	-3.74
$\beta_3$	0.113	15.11	-14.29	0.05	8.55	-11.11
$\beta_1^2$	-	2	-0.003	-0.0019	-0.002	0.0011
$\beta_2^2$	-	-0.19		-0.13	-0.20	0.13
$\beta_3^2$	0.027	×	3.39	-2.20	-4.27	2.15
$\beta_{12}$	-	-	-	-	-	-
$\beta_{13}$	-0.0011	-	-	0.08	-	-
$\beta_{23}$		-1.41	-0.95	-0.69	2	0.58
R <sup>2</sup> adj	58.22	59.45	90.63	83.45	77.76	91.74
EE	0.04	4.58	4.28	2.73	3.64	1.79

βo: constant model; βi: estimated regression coefficient for the main linear effects,  $β^2_i$ : estimated regression coefficient for the quadratic effects,  $β_{ij}$ : estimated regression coefficient for the interaction effects. Subscripts i=1: temperature (°C); i=2: gum Arabic (g/100 g liquidized grapefruit): i=3: carboxymethyl cellulose (g/100 g liquidized grapefruit).  $x_w$ : water content (g<sub>water</sub>/100 g<sub>grapefruit</sub> solids), ε: porosity (%), DMY: dry matter yield (g powder dry matter) g feed dry matter),  $L^*$ : Lightness,  $h^*$ : hue angle and  $C_{ab}^*$ : chroma.

where  $Y_i$  is each one of the response variables and  $x_i$  are the three independent variables, as indicated in Table 2.

#### 3.1. Water content

The water content of the powdered products is related to drying efficiency, playing an important role in its free-flowing behavior and stability during storage, due to its effect on the glass transition and its behavior during crystallization [27]. The water content of the spray dried powders varied between 0.0106 and 0.0828 g water/g powder. Due to the different biopolymers content added to each sample, to make it possible to compare the results of the different powders, they were referred to the grapefruit's own solutes (eq. (1) and (2)). The obtained values (Table 1) were correlated with the independent variables (Table 2). A positive effect of T, GA and CMC on  $x_w$  was observed, along with a negative interaction between T and CMC. As a result, the evolution of  $x_w$  with T and CMC was similar to that shown in Fig 1. At the lowest CMC content, a small  $x_w$  increase occurs when T increases, while at the highest CMC content, a sharp decrease in  $x_w$  is observed when T increases. As the most desirable powdered product would be that with the lowest water content, this will be obtained with a low concentration of GA, an intermediate one of CMC and at high T.



**Figure 1:** Response surface (up) and contour plot (down) of the predicted model for the water content of the spray dried powder, referred to grapefruit's own solutes ( $x_w$ , g water/100  $g_{grapefruit \, own \, solutes}$ ), as a function of carboximethilcellulose content (CMC,  $g/100 \, g_{liquidized \, grapefruit}$ ) and spray-drying temperature (T).

# 3.2. Porosity

The porosity of the powder ranged from 45.3% to 78.2 % (Table 1). It was positively affected by the added solutes, while some negative quadratic effects of GA and a negative interaction between GA and CMC were observed (Table 2). In this way, the highest desired  $\varepsilon$  values will be obtained when a low content of GA and a high CMC content are added (Fig 2).

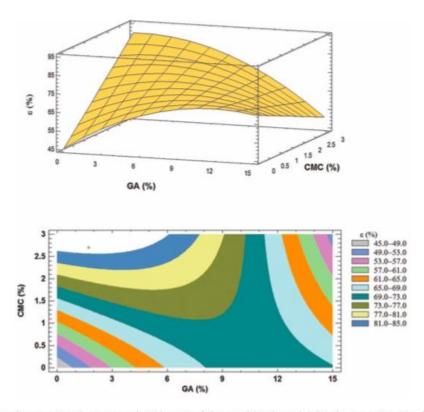
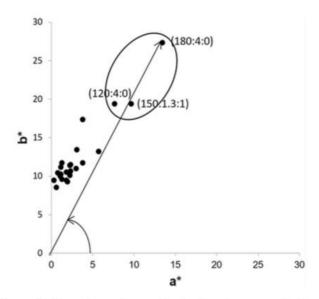


Figure 2: Response surface (up) and contour plot (down) of the predicted model for the porosity ( $\epsilon$ ) of the spray dried powder as a function of GA and CMC content. For the plot, spray drying temperature has been fixed at 150 °C.

### 3.3. *Color*

As far as the color of the different obtained powders is concerned, the L\* values and the chromatic a\* and b\* coordinates are shown in Table 1 and Figure 3, respectively. From a\* and b\* values, the color attributes, hue angle and chroma, were calculated (eq. (5) and (6), Table 1). In Figure 3, the angle described by the sample position to the positive a\* axis is the hue angle and the distance from the sample position to the grid origin (a\* = 0, b\* = 0) is the chroma. As can be seen in Table 1, the L\* values of the different grapefruit powders ranged from 82.3 to 91.5, except for the sample formulated with 4 % GA and dried at 180 °C, which was 66.5, an exceptionally low L\* value. This same sample, together with those obtained at (T = 120 °C, 4 % GA) and (T = 150 °C, 1.3 % GA, 1 %

CMC), also presented, in addition to the lowest L\* values, the lowest hue angle and the highest chroma (Figure 3 and Table 1). The addition of GA to grapefruit increases lightness and decreases a\* [5]. The lower amount of added carbohydrates in these samples could contribute to the observed results, as a lower dilution of the grapefruit pigments occurs. When we visually observed these three samples, despite actually being more orange in color than the rest of them, they were extremely sticky and not a free-flowing powder at all. In fact, they also had the lowest porosity values (Table 1). Taking all these considerations into account, the most desirable color for the grapefruit powder seems to be that which provides a high luminosity and hue angle and a low chroma. In this study, this color is achieved when the liquidized grapefruit is formulated with an intermediate-high GA and intermediate CMC content and is spray dried at an intermediate temperature.



**Figure 3:** Chromatic a\* and b\* grapefruit powders color coordinates. Samples surrounded by a circle are those that presented the lowest hue angle together with the greatest chroma; for them (spray drying temperature:gum arabic concentration:carboximethilcellulose concentration) is indicated. For the sample (180:4:0) hue angle and chroma are indicated with arrows.

#### 3.4. Product yield

Product yield is one of the main indices of the process performance related to its efficiency. DMY ranged from 16.9% to 64 % (Table 1), in the same order as that obtained by other authors working with spray dried pomegranate juice [8] or tamarind pulp [14], for instance. When working with fruits, the low product yield of spray drying is remarkable, which is related to their aforementioned low Tg. In this study the yield was greater than 41 % in only 3 of the 23 spray drying runs carried out, these being the samples

formulated with no CMC and a GA content of over 8 %. This leads to a significant increase in the operational costs, due both to the low process yield and to the need to add a high carrier content in an attempt to increase it. In fact, an economic study carried out in our laboratory, on both laboratory and industrial scales, suggests that the cost of obtaining powdered grapefruit by spray drying is 2.3 times higher than by freeze drying, taking into account all the costs involved in each process [28]. For this reason, it is extremely important to consider the process yield among the response variables when trying to optimize spray drying. In Table 2 and Figure 4, a positive linear correlation with T and GA and a strong negative linear correlation with CMC were observed, together with a negative and positive quadratic correlation with T and CMC, respectively, and a negative CMC-GA interaction. All this behavior leads to the highest product yield being obtained when the liquidized grapefruit is formulated with low CMC and high GA concentration and is dried at an intermediate inlet air temperature. The response surface shown in Figure 4 has been obtained at a spray drying temperature of 150 °C. It maintains the shape but shifts to lower yield values when obtained at 100 or 200 °C, for example. These results highlight the need to study the impact not only of the quantity but also of the type of carrier added. In this sense, CMC does not seem to be a suitable carrier for fruit spray drying. In contrast, the benefit of adding GA has been widely demonstrated [8, 14, 29, 30].

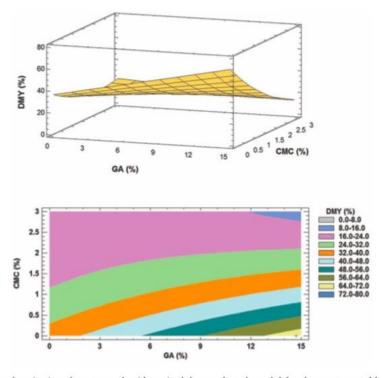


Figure 4: Response surface (up) and contour plot (down) of the predicted model for dry matter yield (DMY, g powder dry matter/ $100 \, g$  feed dry matter) of the spray dried powder. For the plot, spray drying temperature was fixed at  $150 \, ^{\circ} \text{C}$ .

# 3.5. Optimization of temperature and carrier concentration

The final objective of the RSM is to establish the values of the independent variables that optimize the value of the response variables. Optimizing the mix formulation and temperature during spray drying for the purposes of obtaining the best powdered product can pursue different objectives. From the point of view of improving the productive process, it is fundamental to ensure the maximum performance of the process. In our study, when looking for the combination of the independent variables that statistically maximizes the product yield, T = 130.1 °C, 14.44 % GA and 0.035 % CMC were obtained. As can be observed, the temperature of this process is intermediate-lower, the concentration of GA is higher and the CMC is lower than those experimentally considered. According to the results obtained from the different response variables (Table 1), this combination of solutes supposes that the powders obtained would have a high water content and be low in porosity, which is undesirable. If the aim is to obtain a product with the best quality characteristics of those studied, it will be a question of obtaining the one that had the lowest water content and chroma and the maximum porosity, luminosity and hue angle. Statistically, this would be obtained with T = 167.7 °C, 9.30 % GA and 1.28 % CMC. In this case, the GA concentration is intermediate and the CMC concentration is high, implying a low yield. When considering the optimization taking all the variables into account, the conditions would be T = 141.9 °C, 12.82 % GA and 0 % CMC. In this case, the predicted DMY for the process would be 63.97 %. The choice of one or other process conditions will be a function of the final use of the powdered product. From the point of view of the profitability of the process, maximizing the yield will always be desirable. However, considering this parameter alone could only be justified if the final use of the powdered product is as an ingredient in the formulation of other foods. In the same way, prioritizing quality would only be justified if the product were to be offered directly to the consumer, although even in this case it would seem necessary to consider the quality/cost ratio.

#### 4. Conclusions

The results of this study propose adding 12.82 g of GA / 100g liquidized and drying at 141.9 °C, in order to obtain spray dried grapefruit powder with the maximum yield and porosity, the best color and the lowest water content. The use of CMC alone or combined with GA is not recommended. It is not easy to define the best value of the color coordinates and attributes of the powder obtained by spray drying, because what may occur, as happens in the case of the grapefruit, is that the color of the powder that most closely resembles that of the fruit corresponds to an extremely sticky product that could not be considered a free-flowing grapefruit powder. In this study, the best color corresponds to a very luminous powder, with low chroma and orange hue, although much more yellow than red.

# Acknowledgments

The authors thank the Ministerio de Economía y Competitividad for the financial support given through the Project AGL 2012-39103. This mentioned received funding did not lead to any conflict of interests regarding the publication of this manuscript. The authors declare that there is no conflict of interest regarding the publication of this paper.

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