

## INFLUENCE OF KONJAC/SECONDARY GUM BLEND AND SWEETENER ON KONJAC GEL

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This study aimed to investigate the effects of the proportion of konjac in a blend of konjac and secondary gum ( $\kappa$ -carrageenan and xanthan) from 25:75 to 50:50 and the levels of sweeteners (sucrose at 2-20% (w/v) and erythritol-sucralose at 0.2-2.5% (w/v)) on the firmness of konjac gels using response surface methodology. The variation in the proportion of konjac greatly affected the gel firmness with respect to the sweetener. Increasing the konjac proportion increased the firmness of the konjac gels that were co-gelated with either of the secondary gums. At a high konjac proportion, an addition of 2-11% sucrose increased the firmness of the konjac/ $\kappa$ -carrageenan gel but decreased that of the konjac/xanthan gel. The addition of erythritol-sucralose had a small effect on the konjac gel firmness. Predicted models of konjac gels were reliable and can be used to optimize the conditions that provided the greatest firmness. The optimal conditions were found to use the konjac/secondary gum at approximately 50:50 with 17% sucrose or 2.5% erythritol-sucralose for the konjac/ $\kappa$ -carrageenan gel and with 2% sucrose or 0.76% erythritol-sucralose for the konjac/xanthan gel.

**Keywords:** Konjac gel formation,  $\kappa$ -carrageenan, xanthan, sucrose, erythritol-sucralose, response surface methodology.

### INTRODUCTION

Konjac flour, which is a neutral polysaccharide extracted from the tuber of *Amorphophallus konjac* C. Koch, is composed of D-mannose and D-glucose at the ratio of 1.6:1, and these molecules are joined-1,4-linkages (Thomas, 1997). Konjac flour has been shown to function as a thickening and gelling agent depending on the type of food product, such as in jam and jelly or as a fat analogue in low fat meat and bakery products (Akesowan, 2010; Jiménez-Colmenero *et al.*, 2010). In general, konjac flour itself dissolves in water to provide a strong water binding capacity and forms a gel by deacetylation in alkaline solutions (e.g. calcium hydroxide or sodium carbonate) or by combining with secondary gums (e.g.  $\kappa$ -carrageenan or xanthan) (Chin *et al.*, 2009; Delgado-Pando *et al.*, 2011). Apart from its technological properties, konjac also promotes human health as mammalian enzymes cannot hydrolyze its  $\beta$ -linkage structure, which means it has no caloric value. In addition, konjac is considered to be an indigestible dietary fibre; thus, it is useful for weight control, triglyceride and cholesterol reduction, and constipation alleviation (Chua *et al.*, 2010). In Japan, a konjac gel food called 'Konnyaku' is commonly consumed and used as a component of traditional Asian foods (Takigami, 2000).

Jam and jellies are sweetened, gel food products which are composed of high sugar content (50-55%), gums like pectin, gelatin, konjac, juices or fruits and other ingredients. Sugar is typically used for sweetening the product and this makes it popular with consumers, especially children who often like to eat it. The consequent high sugar consumption results in

an adverse risk of developing diabetes, obesity, dental decay or of becoming overweight (Knecht, 1990). One approach to partially or totally replace the sugar content of food products is to use sugar alcohols (which are less sweet than sugar and have a bulking effect) or intense sweeteners (which are much more sweet than sugar and do not have a bulking effect) (Newsome, 1993); however, a mixed sugar substitute blend such as erythritol-sucralose would be an alternative approach. Erythritol-sucralose (98.6:1.4) blend is approximately eight times sweeter than sugar; it provides only 0.18 kcal/g and gives a bulking effect (U-Sing Co., 2012). To achieve an equivalent level of sweetness as when sugar is used, the amount of erythritol-sucralose that is required for the manufacture of jelly having low calories than sugar. This might cause some changes to the gel texture of the low-sugar jelly or other gel food products. Apart from the sweetener, the different characteristics of secondary gums which are used to co-gelate with the konjac to form a gel, would also affect konjac gel characteristics. In general, in the absence of other ingredients, a konjac hydrogel that is formed with  $\kappa$ -carrageenan is more firm and brittle than that formed with xanthan, which produces a more elastic gel (Takigami, 2000; Chin *et al.*, 2009). Thus, the relationship that konjac flour has with secondary gums, in the presence of sweeteners, might be important to consider obtaining a desirable konjac gel texture that corresponds to the consumer expectations of food gel products. In addition, it would be beneficial for the manufacture of low or reduced-sugar gel-based food products made with konjac for

consideration to be a health-promoting food. So far, the information is relatively scarce.

Response surface methodology (RSM) is an effective tool not only to investigate how the variables and their interactions affect the responses but also to optimize process variables (Anderson and Whitcomb, 2005). It was chosen to explore the relationship of the studied variables on the konjac gel texture. Therefore, the purpose of this work was to study the effects of konjac/secondary gums ( $\kappa$ -carrageenan and xanthan) and sweeteners (sucrose and erythritol-sucralose) on the firmness of konjac hydrogels. In addition, the optimal conditions for gel firmness were investigated.

## MATERIALS AND METHODS

**Materials:** The hydrocolloids and sweeteners including konjac flour (Chengdu Newstar Chengming Bio-Tech Co. Ltd, China),  $\kappa$ -carrageenan (MSC5744, MSC Ltd., USA), xanthan gum (KELTROL, CP Kelco, San Diego, CA, USA), sucrose (Carlo Erba Reagenti, Rodano, Italy) and erythritol-sucralose (U-Sing Co. Ltd., Thailand) were used.

**Experimental design:** The total concentration of konjac/secondary gum (containing either  $\kappa$ -carrageenan or xanthan) blend was fixed at a level of 1% (w/v). A central composite rotatable design (CCRD) with combinations coded -1.41, -1, 0, 1, and 1.41 was used. The experimental design of the investigation into the konjac/secondary gum blend and the sweetener is presented in Table 1. The model for the response can be described in the mathematical equation:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_{11}X_1^2 + b_{22}X_2^2 + b_{12}X_1X_2 \quad (1)$$

Where: Y = the response calculated by the model;  $X_1$ ,  $X_2$  = the coded konjac/secondary gum blend and sweetener, respectively;  $b_1$ ,  $b_2$  = linear;  $b_{11}$ ,  $b_{22}$  = quadratic; and  $b_{12}$  is interaction coefficient, respectively.

**Hydrogel preparation:** A mixture of konjac flour and secondary gum was gradually added to distilled water and constantly stirred for 10 mins using a magnetic stirrer. The mixture was heated in a water bath at 90°C for 10 mins. The sweetener was added and the mixture continued to be heated and stirred for a further 20 min. The hot mixture was poured

into gel cups (3 cm diameter  $\times$  2.5 cm height), cooled to room temperature (30°C) and then stored at a refrigerated temperature (10°C) for 24 h prior to analysis.

**Compression test:** Hydrogels were placed in an instrumental room at 23 $\pm$ 2°C for 2 h and then removed from the cups. Firmness (N) was measured in the compression test by a Lloyd texture analyzer equipped with a cutting-type test cell at 250 mm s<sup>-1</sup> crosshead speed. Five samples from each treatment were analyzed.

**Statistical analysis:** Hydrogels from each treatment were prepared in triplicate. The observed response was subjected to an Analysis of Variance (ANOVA) test and multiple regression using the Design-Expert Trial Educational version 8.0.2 software (State-Ease Inc., Minneapolis, MN, USA) (Anderson and Whitcomb, 2005).

## RESULTS AND DISCUSSION

**Statistical analysis and model fitting:** Table 2 shows the firmness of konjac hydrogels subjected to different levels of konjac/secondary gums ( $\kappa$ -carrageenan and xanthan) blend and sweeteners (sucrose and erythritol-sucralose). Firmness values of the konjac/ $\kappa$ -carrageenan/sucrose gel, the konjac/ $\kappa$ -carrageenan/erythritol-sucralose gel, the konjac/xanthan/sucrose gel and the konjac/xanthan/erythritol-sucralose gel ranged from 1.621 to 7.499 N, 5.203 to 14.760 N, 4.284 to 9.617 N and 6.009 to 9.933 N, respectively. The regression analysis revealed that all predictive models ( $Y_1$  to  $Y_4$ ) were significant ( $p < 0.001$ ) and their determination coefficients ( $R^2$ ) ranged from 0.9328-0.9835 (Table 3), which indicates that only 6.72% of the total variations were not explained by the model. In general, the higher  $R^2$  value ( $R^2 > 0.8$ ) represents that the experimental and the predicted values were in a good relationship (Koocheki, *et al.*, 2010). Also, the adj- $R^2$  value of all the models (0.8847-0.9780) was high (Table 3); this therefore shows the significance of the model (Liu *et al.*, 2008). In addition, there was also no significant ( $p > 0.05$ ) lack of fit observed for all models (Table 3). Therefore, it can be concluded that the firmness models of konjac hydrogels under various levels of konjac/secondary gum blend and sweetener were reliable and can be described by the following equations with significant terms:

**Table 1. Two independent variables and their coded and actual values used for analysis.**

Independent variables	Unit	Symbol	Coded levels				
			-1.41	-1	0	1	1.41
Konjac/secondary gum blend	%	$X_1$	25	28.66	37.5	46.34	50
Sweeteners							
- Sucrose	%	$X_2$	2	4.64	11	17.36	20
- Erythritol-sucralose	%	$X_2$	0.25	0.59	1.38	2.17	2.5

$X_1$  = % (w/w) konjac flour in a konjac/secondary gum blend (secondary gums used were  $\kappa$ -carrageenan and xanthan) and  $X_2$  = % (w/v) sweetener.

**Table 2. Experimental conditions and results of firmness of konjac hydrogels subjected to different secondary gums and sweeteners.**

Experimental number	Coded variables		Firmness (N)			
	X <sub>1</sub>	X <sub>2</sub>	Y <sub>1</sub>	Y <sub>2</sub>	Y <sub>3</sub>	Y <sub>4</sub>
1	-1	-1	2.589	5.750	5.451	6.195
2	+1	-1	4.778	11.609	9.617	9.933
3	-1	+1	1.621	5.542	4.900	8.088
4	+1	+1	7.258	13.123	7.450	9.659
5	-1.41	0	1.895	5.203	4.284	6.009
6	+1.41	0	7.499	14.760	7.676	9.112
7	0	-1.41	5.138	9.191	8.895	9.606
8	0	+1.41	5.540	9.799	6.714	9.222
9	0	0	7.218	9.148	8.545	9.597
10	0	0	7.345	9.433	8.556	9.119
11	0	0	6.142	9.195	8.312	9.222
12	0	0	5.977	9.632	8.091	9.585
13	0	0	6.477	9.112	8.112	9.599

X<sub>1</sub> = % (w/w) konjac flour in a konjac/secondary gum blend and X<sub>2</sub> = % (w/v) sweetener; Y<sub>1</sub> and Y<sub>2</sub> = konjac/κ-carrageenan gel with sucrose and erythritol-sucralose, respectively; Y<sub>3</sub> and Y<sub>4</sub> = konjac/xanthan gel with sucrose and erythritol-sucralose, respectively.

**Table 3. Regression coefficients for firmness of konjac hydrogels subjected to different secondary gums and sweeteners.**

Source	Firmness			
	Y <sub>1</sub>	Y <sub>2</sub>	Y <sub>3</sub>	Y <sub>4</sub>
Intercept	20.21	-2.26	-21.55	-16.81
Konjac/secondary gum blend (X <sub>1</sub> )	1.21***	0.30***	1.38***	1.14***
Sweetener (X <sub>2</sub> )	-0.05	-1.96	0.30***	3.10
X <sub>1</sub> X <sub>2</sub>	0.02*	0.06	-7.18E-003*	-0.08*
X <sub>1</sub> <sup>2</sup>	-0.02**	-	-0.02***	-0.01***
X <sub>2</sub> <sup>2</sup>	-0.02*	-	-6.64E-003	-0.02
R <sup>2</sup>	0.9328	0.9835	0.9786	0.9548
Adj-R <sup>2</sup>	0.8847	0.9780	0.9634	0.9226
Lack of fit (p-value)	0.3077	0.0624	0.1473	0.1003

X<sub>1</sub> = % (w/w) konjac flour in a konjac/secondary gum blend and X<sub>2</sub> = % (w/v) sweetener; Y<sub>1</sub> and Y<sub>2</sub> = konjac/κ-carrageenan gel with sucrose and erythritol-sucralose, respectively; Y<sub>3</sub> and Y<sub>4</sub> = konjac/xanthan gel with sucrose and erythritol-sucralose, respectively; \*\*\*Significant at p < 0.001, \*\*Significant at p < 0.01, \*Significant at p < 0.05.

$$Y_1 = -20.21 + 1.21X_{1a}^{***} + 0.02X_{1a}X_{2a}^* - 0.02X_{1a}^{2**} - 0.02X_{2a}^{2*} \quad (2)$$

$$Y_2 = -2.26 + 0.30X_{1a}^{***} \quad (3)$$

$$Y_3 = -21.55 + 1.38X_{1b}^{***} + 0.30X_{2a}^{***} - 0.0072X_{1b}X_{2a}^* - 0.02X_{1b}^{2***} \quad (4)$$

$$Y_4 = -16.81 + 1.14X_{1b}^{***} - 0.08X_{1b}X_{1b}^* - 0.01X_{1b}^{2***} \quad (5)$$

Where: Y<sub>1</sub>, Y<sub>2</sub> = the firmness of konjac/κ-carrageenan gel with sucrose and erythritol-sucrose, respectively; Y<sub>3</sub>, Y<sub>4</sub> = the firmness of konjac/xanthan gel with sucrose and erythritol-sucralose, respectively; X<sub>1</sub> = the proportion of konjac in a konjac/secondary gum blend (X<sub>1a</sub> = κ-carrageenan, X<sub>1b</sub> = xanthan); X<sub>2</sub> = sweetener (X<sub>2a</sub> = sucrose, X<sub>2b</sub> = erythritol-sucralose); \*Significant at p < 0.05; \*\*significant at p < 0.01; \*\*\*significant at p < 0.001.

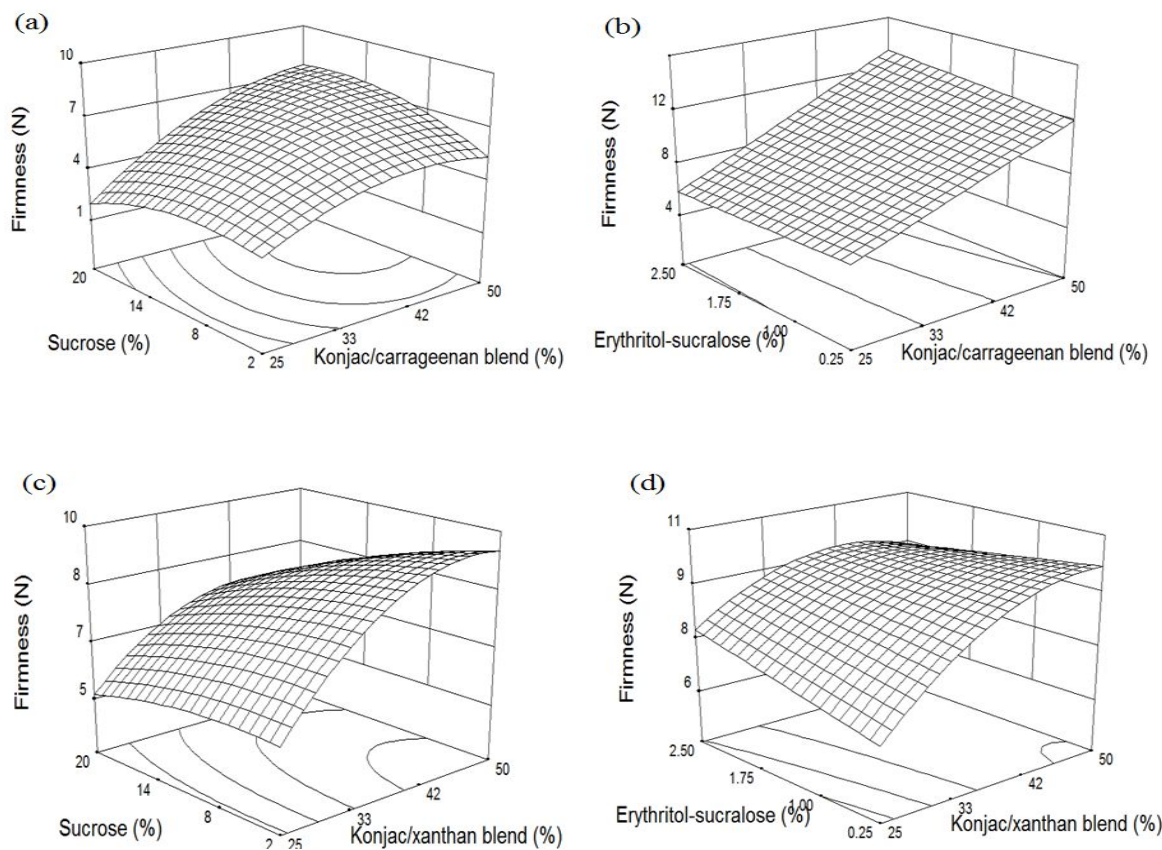
As shown in Eq. 2-3, the konjac/κ-carrageenan proportion was the most important variable affecting the firmness.

Sucrose also had a significant effect on the firmness of the konjac gel formed either with κ-carrageenan or xanthan. The use of erythritol-sucralose showed no effect on the konjac/κ-carrageenan gel (Eq. 3); however, it had an interactive effect with xanthan on the gel firmness (Eq. 5). The effects of the two variables on konjac gel firmness can be better understood by representing the data in three-dimensional or response surface graphs (Fig. 1).

#### Analysis of response surface plots:

##### Effect of konjac/secondary gum blend on konjac gel firmness:

Figure 1 shows response surface graphs generated for konjac gel firmness as a function of secondary gum and sweetener. These graphs reveal the main and interactive effects of the variables, showing a similar ability of secondary gum (κ-carrageenan and xanthan) to increase



**Figure 1. Response surface graphs of gel firmness: (a) konjac/carrageenan/sucrose gel, (b) konjac/carrageenan/erythritol-sucralose gel, (c) konjac/xanthan/sucrose gel and (d) konjac/xanthan/erythritol-sucralose gel.**

mixed gum gel firmness values as increasing the konjac proportion in a konjac/secondary gum blend from 25 to 50% (Fig. 1a-d). In fact, konjac itself does not form a heat-induced gel; nevertheless, it shows a synergistic interaction with these secondary gums (Takigami, 2000). Although the mechanism of intermolecular interactions has not been fully elucidated, the mixed gel might occur when the ordered helical forms of  $\kappa$ -carrageenan associate with konjac molecules through hydrogen bonding (Williams, 2009), whereas konjac attaches to the backbone of xanthan molecules to form a novel sixfold helix, which is an ordered structure (Chandrasekaran *et al.*, 2003; Agoub *et al.*, 2007). As the konjac proportion was increased, a greater number of available konjac/ $\kappa$ -carrageenan or xanthan sites were occupied, which resulted in the generation of a strong gel texture. Moreover, the increase of konjac levels increased the gel strength of konjac gels by increasing of hydrogen bonds formed between konjac molecules (Huang and Lin, 2004). In this investigation, the konjac gel formed with xanthan was more soft and elastic than that formed with  $\kappa$ -carrageenan, which suggests that the characteristics of gel complexes containing interactive polymer networks is

related to the type and the specific properties of each polymer. Consequently, the firmness of the konjac/xanthan gel (soft and elastic gels) (Fig. 1c-d) was more greatly affected by the addition of the sweeteners than the konjac/ $\kappa$ -carrageenan gel (firm and brittle gels) (Fig. 1a-b). The results also suggest that, at the same sweetness level, the firmness value obtained from konjac/ $\kappa$ -carrageenan or konjac/xanthan gels with erythritol-sucralose was higher than that with sucrose. This indicates that the substitution of sugar with erythritol-sucralose is beneficial in reducing the economic costs, as the amount of konjac and secondary gum required to make a low-sugar konjac jelly with gel firmness similar to that made with sugar is lower.

**Effect of sweetener on konjac gel firmness:** Aside from the effect of secondary gums, each sweetener (sucrose or erythritol-sucralose) showed individually effects on konjac gel firmness depending on the type and the concentration of the sweetener that was used. As shown in Fig. 1a, at high konjac levels, increasing the sucrose level from 2 to 11% caused an increase in the gel firmness; afterwards, there was a slight change in firmness as the sucrose level was increased up to 20%. The finding implied that the

manufacture of low-sugar konjac/ $\kappa$ -carrageenan gel in which the level of sucrose reduction was below 11% needed to take an interest on the gel texture. Conversely, the addition of erythritol-sucralose had no effect on the firmness of konjac/ $\kappa$ -carrageenan gel (Fig. 1b). Bayarri *et al.* (2006) showed an increase in the texture of a gel composed of a mixture of  $\kappa$ -carrageenan and locust bean gums with the addition of sucrose. This was attributed to the ability of sucrose molecules to adhere or to retain water. This might increase the number of water molecules, which would favour the interaction between the gum molecules. With regard to erythritol-sucralose, which was used instead of at a range of 0.25-2.5%, as calculated based on equivalent sweetness of sucrose, showing a less amount than that of sucrose used (2-20%). This might be the main reason why erythritol-sucralose showed no effect on the firmness of the mixed gel. Another reason might be due to the difference in water binding capacity, which is much higher for sucrose than erythritol-sucralose.

Conversely, at high level of konjac used, an increase in both sweeteners (sucrose and erythritol-sucralose) decreased the firmness of the konjac/xanthan gels (Figure 1c-d), which was the opposite effect to that of the konjac/ $\kappa$ -carrageenan gel. This finding suggested that the influence of sweeteners on konjac gel firmness would vary depending on which kind of secondary gums were used. The difference in the nature of  $\kappa$ -carrageenan, which can solely form a thermoreversible gel on its own upon cooling, compared with that of xanthan, which cannot form a gel itself but gives a high viscous solution, would be related to the results obtained. With regard to Eq. 4-5, the sweeteners also showed negative interactive effects on gel firmness ( $p < 0.05$ ) when used with xanthan. This indicated that, at lower level of sweeteners, both gels (Fig. 1c-d) showed higher firmness with an increasing konjac proportion in the mixed gum blend; however, the interactive effect reflected a decrease in the mixed gel firmness when higher levels of these two variables were applied. The finding implied the special consideration when the sucrose reduction was made in the konjac/xanthan gel. It should be aware on the decreasing gel firmness and be compensated by increasing gum concentrations or added other ingredients. The greater effect was pronounced for sucrose than erythritol-sucralose, possibly because of the better water holding capacity and the higher amount of sucrose. The decreasing firmness of konjac/xanthan gel with increasing sucrose concentrations can be attributed to various factors together, such as the competition between the sucrose and gums for the available water, the reduced chance of water molecules to interact with the gums molecules, the alteration of water structure and the limitation of gums hydration because of the interaction of gum with sucrose (Knecht, 1990; Totosa *et al.*, 2005; Sharma *et al.*, 2009). However, to gain a more comprehensive understanding further investigation is needed.

**Optimisation and validation:** The analysis of optimal conditions for the maximum gel firmness were as 1% konjac/ $\kappa$ -carrageenan (49.38:50.62) with 16.85% sucrose, showing a firmness value of 7.74 N; 1% konjac/ $\kappa$ -carrageenan (50:50) with 2.5% erythritol-sucralose, showing a firmness value of 13.42 N; 1% konjac/xanthan (47.26:52.74) with 2% sucrose, showing a firmness value of 9.50; and 1% konjac/xanthan (48.47:51.53) with 0.76% erythritol-sucralose, showing a firmness value of 9.95 N. These optimal conditions were suitably validated by the experimental firmness test ( $n = 3$ ), which found to be 7.65, 13.06, 9.24 and 9.82 N, respectively, which shows that the predicted models were reliable.

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