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Effects of different types and concentration of hydrocolloids on mango filling

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Abstract

Hydrocolloid has long been utilized as a thickener to increase the consistency such as fruit jam. In this study, the optimum type (xanthan gum/XG, Arabic gum/AG, guar gum/GG, locust bean gum/LBG, pectin/ PC and carboxymethylcellulose/ CMC) and concentrations (0.2%, 0.4%, 0.6%, 0.8%, and 1% (w/v)) of the hydrocolloid as thickening agents in 'Chok Anan' mango filling was determined using Full Factorial design (108 trials). Physicochemical analyses, including total soluble solid (TSS), pH (acidity), moisture content, and viscosity (Power Law) were conducted. The predicted values for 1.0% PC, and 0.2% XG with the response as TSS (44.9ºBrix), acidity (3.4), moisture content (47.48%) and pseudoplastic behavior (shear thinning) value of n = 0.2917 were similar to experimental optimum values. The optimum type (PC) and concentration of hydrocolloids (1%) of experimental values (TSS= 45.3ºBrix, pH= 3.41) show insignificant different with predicted values. The concentrations (0.2%, 0.4%, 0.6%, 0.8% and 1.0%) and type of the hydrocolloids (XG, AG, GG, LBG, PC and CMC applied in this study had significantly ($p<0.05$) affected the acidity value, TSS, moisture content, and flow behavior index of mango filling. The mango fillings with PC as hydrocolloid may diversify the application of local mango fruits.

Introduction

Hydrocolloids were used in food because of their characteristic such as a thickener, gel, syneresis control, stabilize an emulsifier or suspension, as a coating, and bind water (Sadar, 2004). Hydrocolloids are water-soluble polymers that have the ability to thicken gel or stabilize aqueous systems. The thickening effect of hydrocolloids depends on the type of hydrocolloid used, its concentration, the food system which it used and also the pH of the food system and temperature (Javanmard *et al.*, 2012; Milani and Maleki, 2012; Cropotova *et al.*, 2013). The thickening process includes nonspecific entanglement of conformational disorder polymer chains which is an essentially polymer-solvent interaction (Milani and Maleki, 2012).

Hydrocolloids expressed in food are affected by several factors such as orientation and molecular association, water-binding and swelling, concentration, particle size, degree of dispersion, interaction with other gums and help in improving properties of the filling (Saha and Bhattacharya, 2010; Banerjee *et al.*, 2012; Milani and Maleki, 2012). The addition of gums to food product very often changes the perceived character of food beyond recognition (Uan-on *et al.*, 2008; Cropotova *et al.*, 2015). Certain hydrocolloids or gum usually used as thickening agents in the food industry. For example, gum Arabic from Acacia Senegal tree is said able to reduce viscosity and applied in fruit juice based beverage and soft drinks (Bemiller, 2011; Milani and Maleki, 2012). It influenced the viscosity, body and texture of the food products (Saha and Bhattacharya, 2010). Guar gum and locust bean gum (galactomannans) also act as a thickener, and used in sauces, gravies, and act as moisture retention in the cake, pie, donut and frozen foods (Labuza and Hyman, 1998; Labuza, 2011). Sahin and Ozdemir (2004) also reported that 1% concentration guar gum and locust bean gum contributed to a maximum increase in the consistency of tomato ketchup.

Also, carboxymethyl cellulose (CMC) contribute to high viscosity, but it can reduce by adding electrolytes. For example, the cellulose solution added with sodium chloroacetate reduces the viscosity. The reaction in the solution which may occur the alkali cellulose react with sodium chloroacetate and produce cellulose as by-products (ethers) at low pH condition (Sahin and Ozdemir, 2004, Silva *et al.*, 2013). It is usually applied in salad dressing, gravies, fruit pie fillings and ketchup as thickeners (Koochecki *et al.*, 2009). Nevertheless, pectin is the most common gelling and thickening

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agent in food, and it is extracted from plant cell wall. In food industry; jam and jellies usually used pectin as a thickening agent to change the texture or flow behavior of the final product (Javanmard and Endan 2010). Meanwhile, xanthan gum is stable over a pH range of 2 to 12 which has water binding capacity, and highly soluble in cold or hot water with viscosity behavior fully reversible between 10°C to 80°C (Santos *et al*., 2000; Labuza, 2011). These characters contribute to heat stability, uniform viscosity (Santos *et al*., 2000) and also provide excellent thermal stability for fruit fillings when exposed to high temperature, especially in baking products (Labuza, 2011).

In this study, the mango filling was selected (Chok Anan variety) because it has good texture, flavour and high amounts of antioxidants, mainly b-carotene, phenolic compounds, vitamin C and minerals (Schieber *et al*., 2000) which promoted mango as a suitable raw material for various applications in functional food production (Danalache *et al*., 2015). Neidhrat *et al*., (2002) stated that Chok Anan mango variety has high yield, firmness, and fiber content compared to the other eight mango varieties. However, mango is a highly perishable fruit which can only store without perishing for 4 to 8 days at room temperature and 2 to 3 weeks in cold storage at 13°C with a relative humidity of 85 to 90% (Rimkeeree and Charoenrein, 2014). Thus, this will limit the commercialization of the mango products during its off-season. Akhtar *et al*., (2010) stated that most of the fruit processing industry (in Pakistan) preserves mango pulp for the manufacture of mango products around the year. Mango pulp is not consumed directly, but it is used as fillings for pastries, jams, sauces, fruit juices and drinks (Hussain *et al.*, 2003).

Basu *et al*., (2011) stated that hydrocolloids are used in fruit jams to increase the consistency of the final product to be firm enough to hold the fruit tissue. A study by Croptova and Porpel (2013) revealed that among the pectin, xanthan gum, and carboxymethylcellulose, only low methoxyl pectin show heat-stable properties of fruit fillings (apple filling) within the soluble solid range from 30 to 70°Brix. However, Pichler *et al*., (2012) also stated that addition of the modified starch or hydrocolloids and sugar in the raspberry filling showed the waxy modified starch or guar gum had the greater impact on the filling consistency compared to the Karaya gum and tapioca modified starch.

Therefore, this study aim to optimize and develop improved mango filling with the addition of hydrocolloids (Xanthan Gum, Arabic Gum, Guar Gum, Carboxy Methyl Cellulose, Locust Bean Gum, and Pectin) at different concentrations from 0.2% to 1.0%.

Materials and Methods

Materials

The main ingredient for the mango filling preparation is mango puree (*Mangifera indica* L.) of the Chok Anan mango variety.The maturity index is selected to control the mango puree consistency. Furthermore, other ingredients are castor sugar, glucose syrup, fresh cream (35% fat; Anchor brand), and citric acid (Yummy Bakery Bangi, Malaysia). Hydrocolloids solution with fixed concentration; 0.2%, 0.4%, 0.6%, 0.8% and 1.0% for each type of hydrocolloid i.e. xanthan gum (XG), Arabic gum (AG), guar gum (GG), locust bean gum (LBG), pectin (PC) and carboxymethyl cellulose (CMC). All types of hydrocolloids are purchased from Mylab Scientific Sdn Bhd, Malaysia.

Puree Preparation

Mango (*Mangifera indica* L.) of the Chok Anan variety purchased from Pasar Borong Selangor, Malaysia-based on commercial maturity index (index 3-5 set by Department of Agriculture, Malaysia). The mangoes were washed, peeled and cut. Then the flesh was mashed for puree preparation using a kitchen blender (Model Panasonic PSN-MX800S) then blended for 2 minutes. The puree was packed in the zip-sealed plastic bag (4 cm x 6 cm) and kept in a freezer (-20°C) until further analysis (modified from Ledeker *et al*., 2012).

Filling preparation

Mango fillings consisted of fruit puree, glucose syrup, sugar, fresh cream, citric acid, and hydrocolloids solution (concentration of 0.2%, 0.4%, 0.6%, 0.8% and 1.0%) (Provided by Tolaal Enterprise recipe). Each type of hydrocolloid i.e. xanthan gum (XG), Arabic gum (AG), guar gum (GG), locust bean gum (LBG), pectin (PC) and carboxymethyl cellulose (CMC) was dissolved in 50 ml distilled water (60°C) to obtain 0.2%, 0.4%, 0.6%, 0.8% and 1.0% (w/v) concentration. Whereas the control sample was prepared by mixing all the ingredients (fruit puree, glucose syrup, sugar, fresh cream, and citric acid) without adding any hydrocolloid.

The concentration applied in this study from 0.2% to 1.0% was selected due to the different type of hydrocolloid gave different thickening effect (Sahin and Ozdemir, 2004; Milani and Maleki, 2012). Control (C) used as a reference which consists of mango filling without the addition of hydrocolloids.

The hydrocolloid was added into mango filling with reduction of glucose syrup. For example, 0.2% of hydrocolloid solution will reduce 0.2% from the total amount of sugar and glucose syrup (31%). The type hydrocolloids used in this study were purchased from Mylab Scientific Sdn Bhd, Malaysia.

The filling preparation started with the mango puree thawed overnight at 4°C before mixed with other ingredients and to reduce the browning on the mango puree after thawing. The 46% mango puree was then heated for 5 minutes until the temperature reached 60°C. Later, mango puree was added with the hydrocolloids solution followed by, sugar, glucose syrup (31%) and citric acid (0.5%), and finally fresh cream (22.5%). The puree filling was packed in round plastic container, pasteurized at 80°C for 15 minutes and hermetically sealed. The triplicate sample was prepared for each formulation.

Experimental design

Full factorial design or General Linear Model (GLM) used in this study, and statistical analysis was performed using Minitab Statistical Software (v.17.1.1, Minitab Inc.). Each of formulation was analyzed using ANOVA and GLM to determine which type and concentration of hydrocolloids gave the significant level ($p<0.05$) of total soluble solid, pH value, moisture content and rheological properties of mango filling. The two factors involved in this study were: type of hydrocolloid (6 levels; XG, AG, GG, LBG, PC and CMC) and concentration (5 levels; 0.2%, 0.4%, 0.6%, 0.8% and 1.0%). A total of 108 trials computed by Minitab 17 including the triplicates for each formulation conducted. The optimization using GLM or full factorial design was selected because the number of levels of each factor in this study was more than two. GLM quantified the effect of each response for optimum formulation.

Total soluble solid (TSS), pH, and moisture content

Acidity and sugar content of puree fillings was important to compare between the different type and concentration of the hydrocolloids. Acidity was determined using digital pH meter (Model Mettler Toledo, U.S.A) whereas TSS was measured using a handheld refractometer (Model Atago, Japan). The moisture contents of mango filling formulation were determined using the method of AOAC (2006), with three replicates performed for each sample where the average was calculated. Moisture content (%db) was determined for each sample as the percentage ratio of the weight loss to the initial weight of the sample as in Equation (1) below. The moisture content of mango fillings was analyzed using to compare the moisture content of the mango filling with and without the hydrocolloids additions. The equation for water content calculation is:

$$MC_{db}\ (\%db) = [(W_0 - W_f) / W_f] \times 100 \qquad (1)$$

Where, W_0 = initial weight; W_f = final weight; MC_{db} = moisture content dry basis (%db)

Rheological properties

Rheological properties of mango filling were determined at ambient condition using rheometer (RheoStress 600 Model, Haake, Germany) fitted with a parallel plate geometry (diameter of 35 mm and a gap of 1 mm). The PP35 Ti was used as a probe in this analysis. The measuring device was equipped with a temperature unit (Peltier plate) by programmable water bath with a Universal Temperature Controller (Haake) was used to control the temperature during the measurements. Viscosity test was carried out at shear rate from 5 to $200 s^{-1}$ with a constant temperature of 20°C different from research by Basu *et al*. (2011) with a temperature of 30°C. All mango filling formulation as in Table 1 were measured in triplicates. The viscosity was fitted to the Herschel-Bulkley model where , τ_o, k, and n represent shear stress, yield stress, consistency index, shear rate, and flow behavior index, respectively (modified from Basu *et al*., 2011).

Statistical analysis

The statistical significant ($p<0.05$) of model terms and a fitted regression relationship relating to the response data to the independent variables were analyzed using variance (ANOVA) and GLM. Only significant ($p< 0.05$) terms were included in the final reduced model. The non-significant ($p>0.05$) terms were removed from the initial models and refitted to significant ($p<0.05$) independent variable. This analysis was done to acquire the final reduced model (Mirhosseini *et al*., 2009; Ee 2011). The experimental design matrix, data analysis, and optimization procedure were performed using the Minitab (v.17.1.1, Minitab Inc.) statistical package (Minitab Inc., PA, USA).

Optimization of mango filling formulation and validation procedures

Minitab 17 software was used to define the optimum level of hydrocolloid concentration and type resulted in the overall response goals based on the numerical and graphical multiple optimization procedures. Response Optimizer was used for multiple numerical optimizations to determine

Table 1. Regression coefficients, R^2, adjusted R^2, and probability values for the final reduced models

Regression coefficient	pH value (Y_1)	Total soluble solid (Y_2)	Moisture content (Y_3)	Viscosity (Y_4)
Constant				
b_0	3.490	44.297	49.975	0.9562
Linear				
b_1	-0.0173	-0.891	1.599	0.0402
b_2	0.0191	0.127	-0.426	-0.0039
b_3	-0.0115	-0.629	-0.369	0.1064
b_4	0.0049	0.874	0.338	0.0121
b_5	-0.0101	-0.562	0.626	-0.0546
b_6	0.0149	1.081	-1.767	-0.1003
c_1	0.0535	4.092	-2.329	-0.5612
c_2	-0.0206	0.692	0.257	-0.1010
c_3	-0.0881	-1.364	0.225	-0.0960
c_4	0.1305	-2.303	3.068	-0.1500
c_5	-0.1304	-0.419	0.347	0.2867
R^2	0.5754	0.5132	0.4949	0.6555
R^2(adj)	0.5546	0.4894	0.4702	0.6387
Regression (p-value)	0.000[a]	0.000[a]	0.000[a]	0.000[a]

b_{1-6}, c_{1-5}, d_{1-2}: the estimated regression coefficient for the main linear effects. b_1: Xanthan gum; b_2: Arabic gum; b_3: Carboxymethyl cellulose; b_4: Guar gum; b_5: Locust-bean gum; b_6: Pectin; c1: concentration of 0.2%; c2: concentration of 0.4%; c3: concentration of 0.6%; c4: concentration of 0.8% and c5: concentration of 1.0%

an optimum level of independent variables and responses. According to the optimization procedures, a combined level of concentration and types of hydrocolloids was required to produce several mango filling with desirable fruit filling properties (the lowest acidic value and moisture content, the highest TSS, and viscosity). Verification of final reduced models using t-test was applied to compare the experimental and predicted values for validation procedures. Close agreement and no significant difference must exist between the experimental and predicted values (Ee, 2011).

Results and Discussion

Fitting the general linear model to significant independent variables

All the trial from GLM or Full Factorial model for all formulation with replications for this study were 108 trials. The 108 trials were generated based on the type of hydrocolloid (6 levels; XG, AG, GG, LBG, PC and CMC) and concentration (6 levels; 0%, 0.2%, 0.4%, 0.6%, 0.8% and 1.0%) including control (0% concentration) with triplicates. Therefore, each trial was conducted to determine optimum is filling properties based on target which was the lowest pH, lowest moisture content, highest TSS, and highest viscosity.

The flow properties are the major attributes of fruit filling for consumer acceptance (Wei *et al.,* 2001), whereas the addition of hydrocolloids in this study might result in interaction with starch and modify the flow properties of the fruit filling. Low moisture content in the fruit filling is an important characteristic to prolong shelf-life. It is also dependent on the water activity towards the equilibrium of the components in the fruit filling to avoid diffusion of moisture (Maltinij *et al*., 1993). Therefore, the fitted model was generated to optimize the formulation. The estimated regression coefficients for the response variables, with their corresponding R^2, R^2 (adj), and a p-value of independent variables, were provided in Table 1. Based on Table 1, the full factorial analysis showed significant ($p<0.05$) fitting for all response variables were achieved for all interaction regression models. Therefore, more than 50% of the response variation could be accurately explained as a function of the three independent filling formulation parameters. The response in this experiment was labeled as (Y) whereas (b and c) as the factor.

pH and total soluble solid (TSS) value

Acidity and total solids level are an important quality attribute because it reflects the shelf-life of the fruit filling as well as the flow properties of the filling. The more acidic and high concentration of sugar content show the great potential of an extension of fruit filling shelf-life and contribute to a certain thickness in term of flow properties (Wei *et al*., 2001; Javanmard and Koohikamali, 2011;Javanmard *et al*., 2012). Mean of acidity and sugar content of each filling formulation were analyzed to select the optimized formulation based on the different concentration and type of hydrocolloids applied in this study (Table 2). Thus, the final fitted model shows the pH value had significantly ($p<0.05$) negatively associated with the linear model (Table 1). This result indicated that the higher the hydrocolloid concentration, the greater increment of acidity value (more acidic) of the mango fruit filling. Figure 1(A)

Table 2. Means of pH value, total soluble solid and moisture content for mango fillings with the addition of different types of hydrocolloids and concentrations.

Analysis	Type of Hydrocolloid	Hydrocolloid Concentration				
		0.2%	0.4%	0.6%	0.8%	1.0%
pH	C	3.42±0.06^{abA}	3.42±0.06^{abA}	3.42±0.06^{abA}	3.42±0.06^{abA}	3.42±0.06^{abA}
	XG	3.51±0.01^{aB}	3.42±0.00^{aC}	3.31±0.01^{cE}	3.65±0.01^{aA}	3.37±0.02^{abD}
	AG	3.62±0.06^{aA}	3.43±0.01^{aA}	3.41±0.00^{bA}	3.69±0.07^{aA}	3.45±0.12^{abA}
	CMC	3.64±0.06^{aA}	3.46±0.14^{aB}	3.34±0.01^{cB}	3.69±0.04^{aA}	3.28±0.00^{aB}
	GG	3.56±0.02^{aA}	3.45±0.01^{aB}	3.34±0.01^{cB}	3.63±0.07^{aA}	3.41±0.06^{abB}
	LBG	3.60±0.02^{aA}	3.44±0.01^{aC}	3.51±0.01^{aB}	3.58±0.04^{aA}	3.24±0.01^{bD}
	PC	3.60±0.09^{aAB}	3.50±0.01^{aBC}	3.40±0.01^{bC}	3.72±0.05^{aA}	3.41±0.06^{abC}
Total soluble solid (°Brix)	C	44.7±0.46^{aA}	44.7±0.46^{aA}	44.7±0.46^{aA}	44.7±0.46^{aA}	44.7±0.46^{bA}
	XG	51.4±0.46^{dA}	44.9±0.11^{aB}	46.7±0.58^{abE}	42.0±0.00^{bD}	43.0±0.00^{cC}
	AG	48.8±0.20^{cA}	42.4±0.11^{bcC}	47.5±0.50^{aB}	39.4±0.34^{dD}	45.3±0.58^{bB}
	CMC	48.0±0.00^{cA}	41.8±0.20^{cC}	40.3±0.58^{dB}	41.1±0.11^{cD}	41.7±0.41^{cdCD}
	GG	48.0±0.00^{cA}	42.5±0.11^{bcD}	41.7±0.41^{cdC}	42.2±0.00^{bD}	47.5±0.50^{aB}
	LBG	46.7±0.11^{bA}	42.7±0.64^{bC}	43.0±0.00^{cB}	45.0±0.00^{aB}	40.3±0.58^{dD}
	PC	52.5±0.46^{eA}	41.8±0.23^{bcC}	45.3±0.58^{bB}	42.1±0.11^{bC}	45.3±0.58^{bB}
Moisture content (%)	C	45.65±5.70^{aA}	45.65±5.70^{aA}	45.65±5.70^{aA}	45.65±5.70^{aA}	45.65±5.70^{aA}
	XG	43.23±0.29^{aA}	53.82±0.07^{aC}	53.82±0.07^{aC}	53.41±0.02^{cC}	51.08±0.42^{bB}
	AG	44.70±0.45^{aA}	47.86±0.02^{bB}	47.77±0.01^{bB}	56.12±0.05^{aC}	49.28±0.47^{cA}
	CMC	45.44±0.42^{aA}	47.86±0.03^{bA}	47.86±0.03^{bA}	54.53±0.08^{bB}	51.56±0.84^{bC}
	GG	44.69±0.45^{aA}	51.88±1.19^{aB}	51.90±1.22^{aB}	53.40±0.15^{cB}	46.80±0.34^{dC}
	LBG	47.55±0.29^{aAC}	52.78±0.15^{aC}	52.78±0.15^{aC}	50.08±0.64^{eB}	54.21±0.16^{aA}
	PC	41.60±0.16^{aAB}	46.16±0.37^{bB}	46.16±0.37^{bB}	53.08±0.30^{bC}	47.92±0.87^{cdB}

*C (control without addition of hydrocolloids), XG (xanthan gum), AG (arabic gum), CMC (carboxylmethyl cellulose), GG (guar gum), LBG (locust bean gum), and PC (pectin)

Different small letters (a, b, c, d, e) within a column shows significant different ($p < 0.05$) based on Tukey's test

Different capital letters (A, B, C, D, E) between a column shows significant different ($p < 0.05$) based on Tukey's test

depicts the hydrocolloid concentration gives the main effects for the pH mean value whereas not much different for the type of hydrocolloids. The results were similar to a study by (Wei *et al.*, 2001) who stated that the pH for commercial fruit fillings (apple, lemon, grape, and orange) should be at a lower range of acidity. Nakauma *et al*., (2008) stated the effect may explain predominantly to the isoelectric point of the protein anchor, the electrostatic interactions within and / or between the carbohydrate portions. Thus, the high concentration hydrocolloids create more hydrogen bonds formation in acidic conditions and low water activity in mango puree in order to form mango filling(Javanmard *et al*., 2012).

TSS value of mango filling exhibited significantly ($p<0.05$) negative linear coefficient on the concentration (0.6% to 1.0%) and type (XG, CMC, and LBG) of hydrocolloids according to Table 1. Table 1 showed that there was only linear model were significant for the all term (type and concentration of hydrocolloids) of the coefficient and not dependent on each other. The relationship represents in the main effects plot for pH in Figure 1(B) interpret the main effects for the TSS value was the concentration of the hydrocolloids at 0.2%.

Also, high sugar content in fruit fillings able to prolong the fruit filling shelf life (Young *et al.*, 2003) as the presence of hydrocolloids in both fruit filling systems, had caused the occurrence of competition for the available water between sugars (Croptova *et al.,* 2015). In the available water, there are hydrophilic molecules and the added hydrocolloids, thus inter- chain bonding would be promoted (Nussinovitch, 1997) in the fruit system. Therefore, the physical activities of the water in fruit filling is extremely affected by the added hydrocolloids, but a minor impact was attributed to the total soluble solids content and fruit part (Croptova *et al*., 2015).

The type of hydrocolloids, its concentration, the food system in which it is used and the pH of the food systems are the factors that influence the thickening effect of the food products (Saha and Bhattacharya, 2010, Gundurao *et al.,* 2011). Acidic pH range from 2.6 to 3.5 in commercial fruits filling (apple, blueberry, lemon, and raspberry) (Wei *et al*., 2001) whereas, raspberry filling by (Young *et al*., 2003) showed an acidic range of 3.5 to 3.7. Thus, both studies showed almost similar results as in this study (Table 2) with the acidic condition from 3.31 to 3.72. pH of the filling system did affect the rheological behaviors of ionic hydrocolloids such as pectin (PC) which resulted in decreased of average molecular weight and change of structural (Bemiller, 2011). Based on Table 2, the 1.0% concentration showed the lowest acidity (range between 3.24±0.01 to 3.45±0.12) compared to the other concentration.

The acidic state of bakery fruit fillings ranging from 3.3 to 4.3 has a broad tolerance capability with any hydrocolloids (Young *et al.*, 2003). The acidic condition in a food system enhances the gelation which also depends on the hydrocolloids used because the majority of the carboxylic groups exist in ionic form and promote calcium interactions in the filling added with hydrocolloids (Young *et al.*, 2003). Therefore, in this study, the effect of the different concentration and type of hydrocolloids on pH value showed that the 1.0% LBG had the lowest acidic value compared to the other type of mango filling. However, when compared to the 1.0% PC, there is no significant different with 1.0% LBG. Thus, among the other types and concentrations, the 1.0% concentration may give the lowest acidity value with any the hydrocolloids may select as the optimized formulation.

Table 2 shows all the different concentration (0.2% to 1.0%) and type of hydrocolloids used in this study had significant ($p<0.05$) effect on TSS of mango filling. The randomly dispersed polymer segments in dispersions of solution or solvent may be formed by two or more polymer chains term as 'junction zone'. It is important for the hydroxyl groups in sucrose to form a gel and stabilize the structure or 'junction zone' (Costell *et al*., 2004; Saha and Bhattacharya, 2010). Costell *et al*. (2004) suggested that sugars may create and stabilize the junction zone. This phenomenon took place at lower concentrations of hydrocolloids gels because they found that 0.3% (w/w) k-carrageenan with the increase addition of sugar concentration may increase the viscoelastic system for the gel formation.

The different concentration of XG applied in the mango filling (0.2% to 1.0%) shows significant ($p<0.05$) reduction in TSS value. Thus, it might be due to the polysaccharide chain in XG, which consists of two β-D-glucose units linked through the 1, 4 positions stabilized the solid components (Taylor *et al.*, 2012). Cropotova *et al.*, (2015) stated that the TSS of the apple filling was affected by the addition of the pectin, xanthan gum, and carboxymethylcellulose.

It is suggested that the gel-sol transition occurred in the mango filling is affected by the presence of sugar or glucose. The gel-sol transition happens with the increase in the temperature (heating) which the hydrocolloid molecules become continuous phase (dispersed phase and liquid) and change the hydrocolloid gel to hydrocolloid sol (Sahin and Sumnu, 2006). Nishinari *et al.* (1995) show that sugar did affect the viscoelastic properties of agarose and k-carrageenan gels. Therefore, hydrogen bonds between the polymer and the sugar-OH groups contributed to the solvent structural changes in water as solvent (Costell *et al*., 2004). According to Sudhakar *et al.* (1995), the polymer-polymer bond interaction present in sugar was due to the solvent or water bound with the sugar. Therefore, starch-hydrocolloids interaction existed as the effect from the sugar bond interaction. The addition of hydrocolloids helps the polymer-polymer interactions in enhancing the high TSS and carboxylic groups in ionic form and interact the calcium formation in the mango filling (Wei *et al*., 2001). In this study, PC significantly contributed to the highest TSS (52.5 °Brix at 0.2% concentration) of the mango filling. Both of the high TSS and high acidity creates the thickening factors, extreme condition for the growth of microbial and may extend the shelf life of the mango filling as the moisture transfers compromise quality, stability and safety of the product (Bourlieu *et al.,* 2007).

Moisture content

Li and Nie (2015) stated that hydrocolloids molecules bound water, thus modify the properties of the properties of the food ingredients. Moreover, the hydrocolloid is hydrophilic and able to help retard the moisture loss during short-term storage for 48 hours (Milani and Maleki, 2012). In this study, the moisture content of the mango filling (Table 2) was significantly ($p<0.05$) and positively affected by the linear model of both hydrocolloid types (AG, GG, and PC) and concentrations (0.4% to 1.0%) as represent in Table 1. Thus, the result interpreted that the type and concentration of hydrocolloids' applied in this study affected independently on the moisture content of the mango filling as illustrate in Figure 1 (C). The main effects plot for moisture content indicate that all type of hydrocolloids except PC had the high moisture content mean value while for the concentration the 0.8% gives the highest effects for the moisture content.

McMinn (2002) stated that controlling moisture content can be achieved by removing water or binding it towards the food develops that make it stable to both microbial and chemical deterioration. The result was similar to Javanmard *et al.,* (2012) where the addition of sago starch in mango jam filling which the inter-chain bonding would be promoted because of the competition of the available water between sugars and hydrocolloids presence in the mango jam system.

Thus, the addition of the hydrocolloids decreased the moisture content in the mango filling because of the increased in the solid fraction thus reduces the total moisture evaporation (Javanmard *et al*., 2012). As the effect of the different type and concentration of hydrocolloids on moisture content, Sahin and Feramuz (2005) stated that thickening agents are natural or chemically modified carbohydrates which absorb some of the water presents in the food and eventually making the food thicker.

In this study, the addition of hydrocolloids was necessary because moisture content in the mango puree was very high (40 to 50%) due to the free water available and not binding to any of the mango filling component (Javanmard *et al*., 2012). According to Table 2, 0.2% PC shows significant ($p<0.05$) reduction of moisture content compared to control (C). Additionally, the absence of syneresis with the addition of pectin in the fruit filling as proven (Young *et al*., 2003; Agudelo *et al*., 2014). Thus, the addition of hydrocolloid increases the solid fraction and, therefore, reduces total moisture evaporation, thus resulting in a decrease in the moisture content (Javanmard *et al*., 2012). The similar situation may happen in this study towards the mango filling with the addition of different hydrocolloid solution. Based on Figure 1(C), the PC at all concentration except 0.2% showed significantly higher moisture content when compared to the control. Thus, this may suggest that the type of hydrocolloid such as PC had no significant difference in moisture compared to control (at concentration 0.2% has 41.6% moisture content). This suggested that 1.0% hydrocolloid concentration in the mango filling increased the amount of polymer-polymer in the filling thus, bind with water to form gelation and hold the moisture, causing the moisture content higher than the control.

Viscosity

Table 1 demonstrates the viscosity of mango filling negatively proportional to the linear model of hydrocolloids' type (AG, LBG, and PC) and concentration. The results showed that the type and concentration of hydrocolloids significantly ($p<0.05$) affected the viscosity of the mango filling. The main effects plot for viscosity also illustrated in Figure 1 (D). This finding was similar to Wei *et al.* (2001) who studied the apparent viscosity (n) of the fruit filling (apple, blueberry, raspberry and lemon) varies with type of gum, amount of addition and shear rate.

Table 3 also shows that n, flow behavior index of all types of hydrocolloids (AG, GG, LBG, CMC, and PC) except XG, had increased with the increase of concentrations (0.2% to 0.8%). This finding was similar to Krokida *et al*., (2007) who indicated that flow behavior index increased with the increased of concentration in the mango jam samples within the range of n is 0.27 to 0.99. The increase was due to hydrodynamic forces generated as well as the increase in the alignment of constituent molecules led by the structural breakdown of the molecules (Balestra *et al*., 2011).

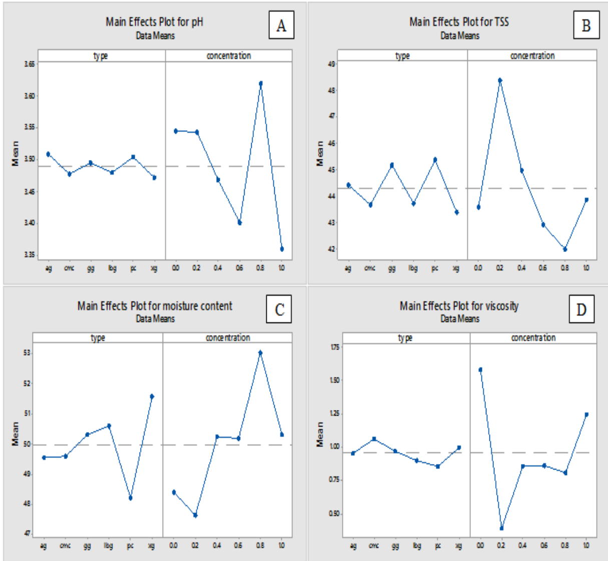

Figure 1. Main effects plot of pH value (A), TSS (B), moisture content (C), and viscosity (D) for mango filling formulations

Based on the Table 3, a few types and concentrations of hydrocolloids showed Power Law behavior with the yield stress, $\tau_o = 0$ (0.2%GG, 0.2%AG, 0.4% CMC and 0.6%XG). Therefore, the mango fillings (0.2%GG, 0.2%AG, 0.4% CMC and 0.6%XG) showed shear thinning properties. This result was similar with Sanchez *et al.* (2009) who had found that guava puree had the shear thinning behavior that fit to the Power Law model. Table 3.5 also shows that the type and concentration of hydrocolloids had significantly ($p<0.05$) affected the viscosity of mango filling. The liquid portion of the fruit filling had shear thinning characteristic based on the Herschel-Bulkley equation. This finding was similar to Wei *et al.* (2001) who found that the apparent viscosity of the fruit filling (apple, blueberry, raspberry, and lemon) varies with the type of gum, the amount of addition and shear rate. According to Wei *et al.* (2001), the flow properties of fruit filling depends on the filling composition and the interaction among the individual components; including acid, starch, sugar gum and water. Bohme *et al.* (2012) stated that the viscosity of the filling was modified during processing due to starch addition. This result was consistent with Uan-on and Senge (2008) who found that the different types of hydrocolloids (GG, XG, and LBG) at concentrations of 0.2% to 1.0%

Table 3. Herschel-Bulkley parameters of mango fillings with the addition of different types of hydrocolloids at different concentrations

Sample	Parameter	Hydrocolloid Concentration (%)				
		0.2	0.4	0.6	0.8	1.0
XG	σ_o	65.32	56.75	0.00	72.29	58.34
	K	0.79	0.34	17.71	0.60	6.96
	n	0.87	0.92	0.34	0.88	0.47
	R^2	0.95	1.0	0.97	0.99	0.95
AG	σ_o	0.00	83.91	61	23.23	45.20
	K	43.12	3.53	0.03	1.74	8.56
	n	0.20	0.48	1.32	0.63	0.47
	R^2	0.98	0.97	0.98	0.98	0.96
CMC	σ_o	55.39	0.00	39.46	47.12	61.95
	K	18.94	30.49	0.95	1.28	4.59
	n	0.30	0.28	0.74	0.70	0.56
	R^2	0.93	0.97	0.99	0.99	0.97
GG	σ_o	0.00	12.49	45.48	30.41	42.69
	K	43.17	26.47	2.19	0.84	28.34
	n	0.20	0.29	0.63	0.72	0.28
	R^2	0.93	0.95	0.97	0.99	0.91
LBG	σ_o	6.56	39.80	39.46	63.13	21.44
	K	36.96	8.77	0.95	2.29	22.29
	n	0.30	0.40	0.74	0.66	0.29
	R^2	0.98	0.96	0.99	0.98	0.95
PC	σ_o	68.62	63.72	59.63	43.82	31.69
	K	34.97	1.02	0.86	1.26	9.88
	n	0.26	0.75	0.77	0.71	0.45
	R^2	0.95	0.98	0.99	0.99	0.96

Herschel-Bulkey parameters: K, consistency coefficient (Pas^n); *n*, flow behavior index, R^2, coefficient of determination and σ_o= yield stress, with the control σ_o= 0.00, K= 46.10, *n* = 0.25, R^2 = 0.95.

affected the flow properties of the pineapple puree.

The flow properties such as non-Newtonian fluid is the major attributes of fruit filling for consumer acceptance (Wei *et al.*, 2001), whereas the addition of hydrocolloids in this study might result in interaction with starch and modify the flow properties of the fruit filling. Low moisture content in the fruit filling is also an important characteristic to prolong shelf life. Viscosity also depends on the water activity towards the equilibrium of the components in the fruit filling to avoid diffusion of moisture (Maltinij *et al.*, 1993). Similarly, Sahin and Ozdemir (2004) stated that the consistency index and the apparent viscosity had increased with the addition of hydrocolloids (taraganth gum, GG, CMC and LBG) with the increase in their concentrations (0.5% and 1.0%).

The entire mango fillings added with hydrocolloids had non-Newtonian (Herschel-Bulkley model with K>0, 0<n<∞, yield stress value, >0) and shear thinning (pseudoplastic) fluids power law (shear- thinning when K>0, 0 < n <1 and yield stress value= 0) characteristics. Similar results were also reported for raspberry cream filling with hydrocolloids addition (karaya gum and GG at 0.05% concentration) by Pichler *et al*. (2012).

The addition of PC in mango filling affected the consistency and decreased the fluidity as shown in Table 3. Galkowska *et al*. (2013) showed that the rheological and textural of starch-pectin-sucrose systems were affected by high methoxyl pectin and sucrose because the starch-pectin-sucrose gels exhibited better structuring ability compared to modified potato starch alone.

Moreover, the addition of mango puree improves the structure of the fillings. Agudelo *et al.* (2014) also concluded that the peach puree developed the structure of the fillings and the baking tests with better performance (the heat stable filling and stay inside the crust) compared to control. Most of the manufactured pectin solutions behave like shear thinning, and is dependent on the raw material as well as the extraction conditions. Otherwise, any alterations in the increase of pH and ionic strength can result in changes in the viscosity of the pectin solutions (Shon and Yoo, 2006).

The yield stress value was calculated using Herschel-Bulkley model which $\sigma = \sigma_o + k\gamma^n$ (Sun and Gunasekaran, 2012). Flow curves exhibited shear thinning characteristics (*n*<1) at all concentrations studied. The flow behavior index, n, informed on the deviation from the Newtonian flow, for which n = 1. Parameter for all mango fillings had showed that the fillings were pseudo plastic, shear thinning liquids (shear-thinning when K>0, 0 < *n* <1 and yield stress value, σ_o = 0) (Herschel-Bulkley model with K>0, 0<n<∞, yield stress value, σ_o>0) (Steffe,

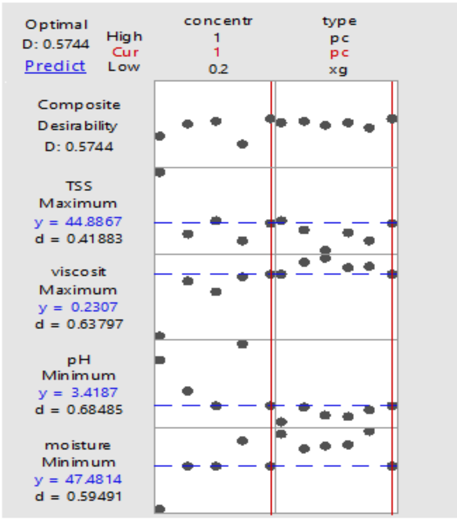

Figure 2. Optimization plot of mango filling formulation

1996; Schramm, 1998; Sikora *et al*., 2007). Thus, the Herschel-Bulkley model fitted the viscosity of mango fillings as in Table 3. The mango filling exhibited the characteristics of the Herschel-Bulkley model because the presence of a yield stress (σ_o) which represents a finite stress is required to achieve the flow. Below is the yield stress of a material with sold-like characteristics which stores energy in small strains and does not level out under the influence of gravity to form a flat surface (Steffe, 1996).

Optimization of Mango Filling Formulation

Optimization of GLM prediction was based on the pairwise comparisons and the main effects plot for each response. The targeted value for pH was low, which reflects the acidic condition. The acidic condition helps to slow the microbial growth in the mango filling. Figure 1(A) shows that the 1.0% XG had the lowest mean of pH value.

Referring to Figure 1(B), Main effect plot indicates that means of TSS value for the mango filling formulation was significantly ($p<0.05$) high with the addition of PC. To extend the shelf life of the mango filling, the polymer-polymer bond interaction which contains sugar to bind with solvent or water to reduce the moisture. Figure 1(B) presents significantly ($p<0.05$) the highest mean of TSS with 0.2% concentration. The main effects plot for moisture content (Figure 1(C)), shows the lowest moisture content also at 0.2% PC. The viscosity of the mango filling present significantly highest viscosity as a thick and apparent viscosity. Figure 1(C) shows 1.0% GG had the highest mean of apparent viscosity besides control. From Figure 2 showed that 1.0% PC (after optimization) contributed to the maximum TSS value at 44.88 °Brix, maximum viscosity of 0.2307, minimum pH value (acidic) 3.41, and minimum moisture content was 47.5%.

Conclusion

Based on the optimization of the mango filling formulation using GLM, formulation 1.0% PC was selected. The optimized 1.0% PC in mango filling has the optimum condition of low pH (acidic), high TSS, high viscosity and low moisture. GLM indicated significant value ($p<0.05$) in all reduced models with a high coefficient of the determined value ($R^2>0.80$). The empirical equation had been developed with validation and prediction of each response variable studied. Thus, it indicates that the properties of newly formulated mango filling could be obtained by addition of hydrocolloids. It is suggested that the linear term of different hydrocolloid type was the most significant ($p<0.05$) factor affecting the response variable i.e. pH value, TSS, moisture content and viscosity (physicochemical properties). Therefore, the different type of hydrocolloid type was found to be the most important variable in affecting the physicochemical properties of the mango filling. This study may help diversify the application of mango puree as filling with acceptable shelf life, the rheological and physical properties.

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