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Analysis of Extruded Pectin Extraction from Apple Pomace by Response Surface Methodology

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Abstract To extract apple pectins, apple pomace (AP) was extruded under 14 different conditions of screw speed (250-350 rpm), feed rate of 30-40 kg/hr, and 20-30% moisture content using twin-screw extrusion. Response surface methodology (RSM), based on three variables by three-level factorial design, was employed to investigate effects of screw speed, feed rate, and moisture on dependent variables of extrudates, soluble dietary fiber (SDF), yield of anhydrogalacturonic acid (Y_{AGA}) representing pectin, and intrinsic viscosity ([η]). Second order models were used to generate three-dimensional response surface for dependent variables, and their coefficients of determination (R^2) ranged from 0.96 to 0.99. Moisture content showed highest effect on solubilization of AP.

Key words: extrusion, apple pomace, response surface methodology

Introduction

Apple pomace (AP) is the primary by-product of the apple juice industry. Attempts have been made by numerous researchers to utilize AP as value-added products including bio-fuels, citric acid, edible fibers, and fertilizers (1, 2). Presently, AP is also employed as a raw material for commercial pectin production (3). Most soluble fractions of apples are separated as juice, whereas a majority of AP exists as insoluble component consisting of macromolecular cell wall materials such as cellulose, hemicellulose, and pectins. Thus, AP does not impart nutritional benefits as a soluble dietary fiber (4, 5). Accordingly, to obtain AP as a soluble dietary fiber, it is necessary to disintegrate the insoluble intermolecular network of AP.

Extrusion of foods is a technology, which enables food industries to process and market a large number of products of varying size, shape, texture, and taste. An extruder simultaneously performs, among others, mixing, cutting, crushing, pressing, expanding, drying, and sterilizing, etc(6). Extrusion technology is widely used in cereal and porous snack preparations (7-10). In particular, this process provides shearing effects concomitantly at high temperature and high pressure (11). Extrusion has been also applied to the modifying of the rigid cell wall structure (12).

Mechanical disintegration of by-products of fruits and vegetables by extrusion increased functional properties such as solubility, swelling power, water-holding capacity (13-15). Ralet and Thibault (16) also reported that highly methylated pectins with long side-chains were produced from lemon cell walls by twin-screw extrusion. In a previous report (17, 18), twin screw extruder was employed to disintegrate AP to facilitate the production of apple pectins by water extraction.

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Response surface methodology (RSM) is a collection of mathematical and statistical techniques useful for analyzing problems, in which several independent variables influence a dependent variable or response. This method is a powerful tool in experimental product development design and has been applied to optimize several food processing operations (15, 19-21).

The purpose of this research was to use RSM as a modeling tool to determine the effects of the processing variables of extrusion such as screw speed, feed rate, and moisture content on the extraction of pectin from AP.

Materials and Methods

Materials AP, supplied in a dried form from a commercial apple juice plant (Kyongbuk, Korea), was ground in a hammer mill to pass through a 80-mesh screen and stored at -10°C. The proximate compositions of AP were: moisture, 11.0%; crude protein, 3.5%; crude fat, 4.7%; ash, 1.8%; carbohydrate, 71.2%.

Extrusion of apple pomace Extrusion was performed using a corotating and intermeshing-type twin-screw extruder (Biex-DNDL 44, Bühler Brothers Co., Horgen, Switzerland) with 20:1 of length to diameter ratio and equipped with a K-tron twin screw volumetric feeder (K-tron Corp., Pitman, NJ, USA). The extruder was designed with modular 176 mm barrels and bored with two 44-mm diameter holes, and the diameter of the die nozzle was 2 mm. The barrel temperature was maintained at 25, 50, 90, and 170°C from the feeding zone to the die section throughout the experiments. AP was extruded under 14 controlled conditions of varying severity (Table 1) based on the fractional factorial experimental design for three variables at three levels (22).

Dietary fiber Dietary fiber was assayed by the method of Prosky *et al.* (23) using a commercial assay kit (Sigma

Table 1. Expermental design for extrusion of apple pomace

Exp. No.	Independent variables[1]		
	X_1	X_2	X_3
1	250	40	25
2	250	35	30
3	300	30	30
4	250	40	20
5	300	40	30
6	350	30	25
7	250	30	25
8	350	35	30
9	300	35	25
10	300	35	25
11	350	40	25
12	300	30	30
13	350	35	20
14	300	40	20

[1]X_1: screw speed(rpm), X_2 : feed rate (kg/hr), X_3 : moisture content (%)

Chemical Co., St. Louis, MO, USA). Depending on the solubility, dietary fiber was divided into two distinct components, i.e., soluble dietary fiber (SDF) and insoluble dietary fiber (IDF). Total dietary fiber (TDF) was the sum of SDF and IDF.

Fractionation of water soluble polysaccharide (WSP) The extruded AP (10 g) was solubilized in 200 mL of distilled water for 1 hr at 25°C, and centrifuged for 10 min at 6,500 × g. Supernatant was filtered through a filter paper, and 800 mL of isopropanol was added to the filtrate. Precipitate was washed with isopropanol and acetone, and dried at room temperature. Dried WSP was solubilized with distilled water to make 1% solution, centrifuged for 10 min at 6,500 × g, and freeze-dried. Yield of WSP was expressed as concentration (g/dL) in the extraction medium.

Measurement of anhydrogalacturonic acid Anhydrogalacturonic acid (AGA:MW=176), the backbone of pectins, was employed as a measure of the amount of pectin in water-soluble polysaccharides (WSP). AGA content was determined colorimetrically using the *m*-hydrodiphenyl method of Bluemen-krantz and Abor-Hansen (24). The standard was prepared using galacturonic acid monohydrate (MW=212; Adldrich Chemical Co., Milwauke, WI, USA), which was corrected to AGA by multiplying with 176/212.

Determination of intrinsic viscosity Intrinsic viscosity of WSP was measured using a Cannon-Fenske glass capillary viscometer (size 50, Cannon Instrument Co., Pennsylvania, PA, USA) at 25°C. Known amounts of WSP were dissolved in distilled water. Solutions were filtered through a 0.45 μm Millipore filter to remove dust and insoluble impurities. Subsequently, 10 mL solution was pipetted into the capillary viscometer, equilibrated in a 25°C water bath for 30 min prior to measurements.

Specific viscosity (η_{sp}) and intrinsic viscosity ([η]) were determined as follows:

$$\eta_{sp} = (\eta - \eta_s)/\eta_s$$

$$[\eta] = \lim_{c \to 0} \frac{\eta_{sp}}{C}$$

where η is the solution viscosity, η_s is the solvent viscosity, and C is the solution concentration.

Statistical analysis In selecting the conditions for the extruder operations, independent variables including screw speed (X_1), feed rate (X_2), and feed moisture content (X_3) were incorporated in a three-level factorial experimental design (Table 1). Dependence variables, SDF, yield of anhydrogalacturonic acid (Y_{AGA}) representing pectin, and [η], were expressed individually as a function of the independent variables.

Data were analyzed by multiple regression to fit the second order (quadratic) polynomial regression model to determine the relationships between the process and functional variables. Response surface methodology (RSM) was applied on the experimental region defined. Experimental data analysis and estimated response surface were performed with multiple regression analysis and surface plotting using SAS (static analysis system). Regression models with linear and quadratic terms were used to create three-dimensional response surface.

Results and Discussion

Soluble dietary fiber (SDF) Statistical analysis of response surface model indicated that changes in SDF were affected the most by the moisture content, followed by feed rate, whereas the screw speed showed no significant effect (Table 2). Regression model accounts for 98% of the total variation (P<0.01) in the SDF values. The regression coefficients of model for SDF as a function of the independent variables are shown in Table 3. Linear term of screw speed, feed rate, and moisture content significantly contributed to the model.

Response surface for SDF as a function of feed rate and moisture content is presented in Fig. 1. SDF was more affected by the moisture content than the feed rate, and decreased with increasing moisture content, due to the fact that AP is less affected by the shearing force at high

Table 2. Analysis of variance showing effects of treatment variables and interaction effects on response variables

Source	Degree of freedom	Sum of squares			
		SDF	Y_{AGA}	[η]	$(Y[\eta])_{AGA}$
Model[a]	9	26.11[3]	0.05[2]	10.43[3]	0.83[4]
Linear	3	20.47[3]	0.04[3]	9.47[3]	0.71[4]
Quadratic	3	3.90[2]	0.01[1]	0.59[1]	0.09[3]
Cross	3	1.73[1]	0.0005	0.38	0.04[2]
R^2		0.98	0.96	0.99	0.99
X_1	4	1.83	0.001	0.63[1]	0.05[2]
X_2	4	8.20[2]	0.01	0.54	0.03[2]
X_3	4	18.35[3]	0.04[3]	8.66[4]	0.70[4]

[1]Significant at 10% level; [2]Significant at 5% level; [3]Significant at 1% level; [4]Significant at 0.1% level
[a]Response surface model : $Y = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_3X_3 + \beta_{11}X_1^2 + \beta_{12}X_1X_2 + \beta_{22}X_2^2 + \beta_{13}X_1X_3 + \beta_{23}X_2X_3 + \beta_{33}X_3^2$ where Y is dependent variables, X_1 is screw speed, X_2 is feed rate and X_3 is moisture content.

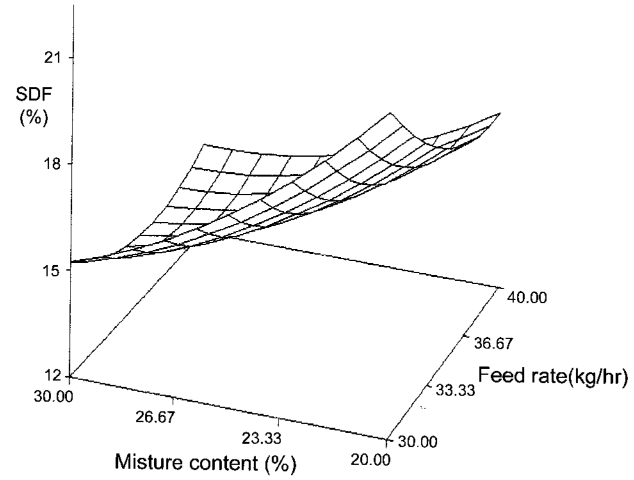

Fig. 1. Response surface plot showing the effect of moisture content and feed rate on the SDF.

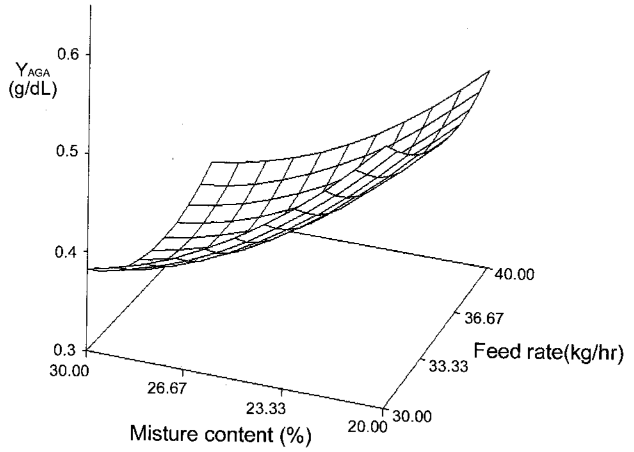

Fig. 2. Response surface plot showing the effect of moisture content and feed rate on the Y_{AGA}.

moisture content. In addition, SDF greatly decreased with increasing moisture content at low feed rates, whereas slightly decreased at high feed rates, because decreasing the residence time of the material within the extruder causes a decrease in the degree of shearing (19, 25).

Yield of anhydrogalacturonic acid(Y_{AGA}) Y_{AGA} was affected by the moisture content (P<0.01), but not by the screw speed and feed rate (Table 2). The regression model conferred a high coefficient of determination (R^2=0.96, P<0.05). Y_{AGA} decreased from 0.53 to 0.36 g/dL when the moisture content increased from 20 to 30% (Fig. 2).

Intrinsic viscosity([η]) Intrinsic viscosity depends primarily on the molecular weight of polymer under the same conditions of chain rigidity and solvent types (25). Regression model accounts for 99% of total variation (P<0.01), and intrinsic viscosity was affected by the moisture content (P<0.001) (Table 2). The linear term of screw speed and moisture content contributed significantly (P<0.05) to the model (Table 3).

Relationship between the moisture content and screw speed on the intrinsic viscosity is presented in Fig. 3.

Table 3. Summary of response surface models; regression coefficients of second order polynomials for response variables[a]

Coefficient	Response variables			
	SDF	Y_{AGA}	[η]	$(Y[\eta])_{AGA}$
β_0	176.37[3]	4.780[2]	-48.79[2]	-10.29[3]
β_1	-0.223[3]	-0.005	0.088[1]	0.024[2]
β_2	-4.313[3]	-0.129[2]	0.868[1]	0.022
β_3	-3.497[2]	-0.096	1.573[2]	0.531[3]
β_{11}	0.0002[2]	-0.000004	-0.00005	-0.00001
β_{12}	0.001	0.00003	-0.001[1]	-0.0003[2]
β_{22}	0.041[2]	0.002[2]	-0.007	0.001
β_{13}	0.001	0.00004	-0.001[1]	-0.0004[3]
β_{23}	0.033[2]	0.0003	-0.003	0.00003
β_{33}	0.031[2]	0.001	-0.018[2]	-0.007[3]

[1]Significant at 10% level; [2]Significant at 5% level; [3]Significant at 1% level

[a]Response surface model : $Y = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_3X_3 + \beta_{11}X_1^2 + \beta_{12}X_1X_2 + \beta_{22}X_2^2 + \beta_{13}X_1X_3 + \beta_{23}X_2X_3 + \beta_{33}X_3^2$ where Y is dependent variables, X_1 is screw speed, X_2 is feed rate and X_3 is moisture content.

Moisture content was the major factor affecting the intrinsic viscosity, while the effect of screw speed on intrinsic viscosity was much less. As the moisture content increased, the intrinsic viscosity increased almost linearly irrespective of the changes in the screw speed. This is in contrast to SDF and Y_{AGA}, which decreased with increasing moisture content (Fig. 1 and 2). Hwang *et al.* (18) reported that extrusion conditions, increasing the yield of WSP, decreased the intrinsic viscosity and the molecular weight of pectin. Anisworth *et al.* (25) and Kim *et al.* (27) reported that an increase in moisture content reduced the viscosity of extruded materials. This difference might be due to the physical properties of raw materials such as chemical composition and particle density, which are strongly related to the water adsorption capacity and viscosity of dough during extrusion.

Yield of AGA × intrinsic viscosity [$(Y[\eta])_{AGA}$] From the industrial viewpoint, the primary goal of pectin extraction is to maximize the yield and molecular size of pectin. However, as clearly demonstrated by the experimental data, the opposite trends of these two objective functions, yield and intrinsic viscosity, make it difficult to optimize the extrusion conditions (16). Thus, a dimensionless optimization parameter, i.e. yield of AGA × intrinsic viscosity or $(Y[\eta])_{AGA}$, could be used as the objective function for optimization.

The regression model accounts for 99% of the total variation and significance at the 0.1% level in the $(Y[\eta])_{AGA}$ values (Table 2). $(Y[\eta])_{AGA}$ was affected by the moisture content (P<0.001), screw speed (P<0.05), and feed rate (P<0.05), of which the linear effect of moisture content was founded to be the most significant (P<0.001) (Table 3). Response surface for $(Y[\eta])_{AGA}$ as a function of moisture content and screw speed is presented in Fig. 4. $(Y[\eta])_{AGA}$ increased linearly with increasing moisture content, whereas the screw speed showed a minor effect on this dimensionless parameter.

In conclusion, Response surface methodology (RSM)

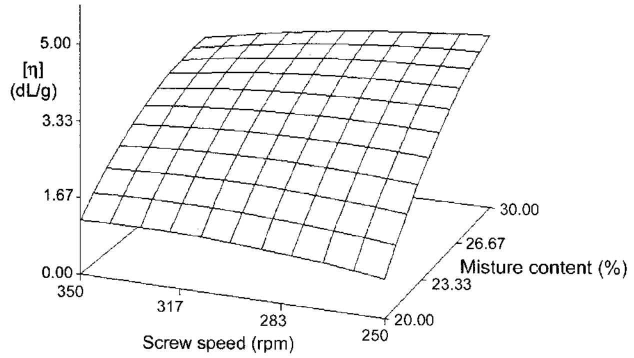

Fig. 3. Response surface plot showing the effect of moisture content and feed rate on the [η].

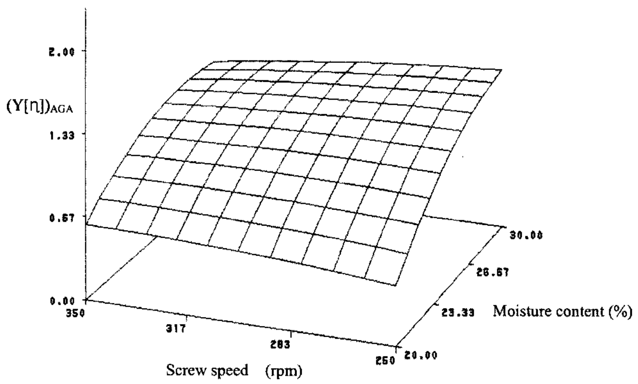

Fig. 4. Response surface plot showing the effect of moisture content and screw speed on the $(Y[\eta])_{AGA}$.

was employed to investigate the effects of screw speed, feed rate, and moisture content on the dependent variables involved in the extraction of pectin from apple pomace. Of these three process variables, the moisture content was the most significant factor affecting the solubilization of apple pomace by extrusion.

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