Response Surface Methodology (RSM) and Artificial Neural Networks (ANNs) Pectin Extraction from Banana Peel: A Modeling and Optimization Approach

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ABSTRACT: *In the present study, the extraction of pectin from the banana peel (Musa sp.) was optimized using an artificial neural network and response surface methodology on the yield and degree of esterification obtained using microwave-assisted extraction methods. The individual, quadratic, and interactive effect of process variables (temperature, time, the liquid–solid ratio, and pH) on the extracted pectin yield and DE of the extract were studied. The results showed that a properly trained artificial neural network model was found to be more accurate in the prediction as compared to the response surface model method. The optimum conditions were found to be the temperature of 60ºC, extraction time of 102 min, the liquid–solid ratio of 40 % (v/w), and pH of 2.7 and within the desirable range of the order of 0.853. The yield of pectin and degree of esterification under these optimum conditions were 14.34% and 63.58, respectively. Temperature, time, liquid–solid ratio, and pH revealed a significant ($p < 0.05$) effect on the pectin yield and degree of esterification. Based on the value of methoxyl content and degree of esterification the extracted pectin was categorized as high methoxyl pectin. Generally, the findings of the study show that banana peel can be explored as a promising alternative for the commercial production of pectin.*

KEYWORDS: *Banana peel; Pectin; Artificial Neural Networks; Response surface methodology; microwave-assisted; Extraction.*

INTRODUCTION

The cultivated desert banana and plantain (Musa sp.) are considered some of the most important food crops for the tropical and subtropical regions and play important role in food security and the economy (*Waghmare* and *Arya*, 2016). In Ethiopia, the banana is considered the most popular fruit crop that is broadly grown and consumed. It covers about 60 % of the overall fruit area, approximately 68 % of the entire fruits produced, and about 38 % of the total fruit-producing farmers (*Woldu et al.,* 2015). Bananas are most widely consumed as raw, and processed into products such as banana flour, chips/crackers, and puree. Banana fruit peels constitute about 30% of the fruit and represent an environmental problem due to their large nitrogen and phosphorus contents as well as their high

water content, making them highly susceptible to microbial degradation (*Oliveira et al.,* 2015). Utilization of banana peels as a source of high-value compounds like pectin (*Happi Emaga et al.,* 2008), cellulose nanofibers, and phenolic compounds is interesting from an economic point of view and an environmental perspective (*González-Montelongo et al.,* 2010).

The most generally used method of extracting pectin is by strong acid concentration but this method is corrosive, adverse to the environment, has a high cost for treating acidic waste, and may be a potential threat to health (*Vriesmann, Teófilo,* & *Lúcia de Oliveira Petkowicz*, 2012). Microwave-assisted extraction with a weak organic acid is effective in pectin extraction and pectin properties (*Phaiphan et al.,* 2019). The effect of process parameters on pectin yield during extraction from banana peels has been studied by several researchers (*Khamsucharit, Laohaphatanalert, Gavinlertvatana, Sriroth,* and *Sangseethong*, 2018; *Phaiphan et al.,* 2019), however, to the best of our knowledge, there was no report on the microwave-assisted extraction of pectin and Degree of Esterification (DE) from banana peel by coupling Artificial Neural Network (ANN) and Response Surface Methodology (RSM). The RSM is a statistical mathematical tool that is widely employed to examine multiple regression analysis using quantitative data obtained from appropriate experiments to determine and simultaneously solve multivariant equations. ANN is a powerful mathematical method suitable for modeling and simulation of various processes in real engineering applications.

Pectin is considered one of the most valuable products which can be primarily extracted from apple pomace, citrus peel, guava extract, sugar beet, and sunflower heads. Pectins are widely used as a functional ingredient within the food industry, pharmacy, and cosmetic manufacture thanks to their ability to form aqueous gels, and dispersion stabilizers (*Mohamed*, 2016). Generally, two types of pectin are available in nature such as high methoxyl pectin (HMP; greater than 50% DE) and low methoxyl pectin (LMP; below 50% DE) forms gel after heating in sugar solutions at a concentration above 55% and pH below 3.5.

In the present work, RSM and ANN-linked genetic algorithm-based models have been developed to predict the relationship between the input variables and the output variables. Subsequently, the result predicted by the ANN

and RSM techniques were compared statistically to the coefficient of determination (R^2), Root Mean Square Error (RMSE), Mean Absolute Error (MAE), standard error of prediction (SEP%), and Absolute Average Deviation (AAD%) based on the validation data set for their predictive and generalization capabilities. An effective RSM model and a feed-forward neural network on back-propagation were developed by utilizing the experimental data, and the efficiency of both models was compared. Therefore, this study was conducted to investigate the effect of extraction conditions namely, temperature, extraction time, Liquid-Solid Ratio (LSR), and pH on yield and DE and to optimize these conditions by employing ANN and RSM. In addition to this, pectin was extracted from banana peel by using microwave-assisted methods to perform the chemical characterization of the extracted pectin in order to evaluate its suitability in food industries.

EXPERIMENTAL SECTION
Materials
The banana peel was collected from some selected hotels, juice processing houses, and restaurants in Jimma town, Ethiopia. All chemicals used for the extraction process were of analytical reagent grade.

Raw material preparation
The fresh banana peels were segregated according to their type and chopped into approximately 1cm^2 pieces using a stainless steel knife for easy drying and washed with water three times. Sample drying was carried out in an oven at 60°C for 48 hours to obtain easily crushable material. The dried peel was ground by a Wiley mill and then screened to pass through a sieve size of 60 meshes and packed in an airtight, moisture-proof bag at room temperature and ready for the extraction process.

Pectin extraction using Microwave-Assisted Extraction (MAE) methods
In this study, pectin was extracted according to the methodology proposed by *Li et al.* (2012) with a few modifications. Depending on the LSR specified in the experimental design, dried banana peel powder was subjected to extraction by adding 0.05 M of hydrochloric acid (HCl) of different pH. For MAE extraction, the mixture was extracted by microwave with a power of 300 W

Table 1: Standard Methods used for physicochemical characteristics of extracted pectin.

Parameters	Methods
Equivalent Weight	(Ranganna, 1995).
Methoxyl Content	(Ranganna, 1995)
Total Anhydrouronic Acid Content	(Mohamed S & Z., 1995)
Degree of Esterification	(*Liew et al.,* 2014a)
Moisture Content Determination	AOAC Method 934.01(AOAC,2005)
Ash Content Determination	AOAC, (2005)

for the four independent variables. After that, the sample was centrifuged (4500 rpm for 30 mins) and the supernatant was precipitated by 96% of ethanol (1:1 the supernatant to ethanol, V/V). The mixture was incubated for 15 hrs at room temperature. Thereafter, the precipitated pectin was filtered using nylon/muslin cloth, which was followed by washing three times with 95% of ethanol and 50% of acetone to get rid of sugars, such as monosaccharides and disaccharides (*Minkov et al.*, 1996). Finally, the wet pectin extract was dried at 40°C in a hot air oven overnight to remove the moisture until its weight remained constant and ground into powder and the percentage yield of pectin was determined according to *Ranganna* (2004).

$$\text{Yield of Pectin (\%)} = \qquad\qquad\qquad (1)$$

$$\frac{\text{Amount of extracted pectin (g)}}{\text{Initial amount of bababa peel (5g)}} \times 100$$

Analytical methods

A sample of dried banana peel pectin was subjected to a quantitative test in order to determine its physicochemical characteristics. From the results obtained, the optimal conditions that gave the optimum yield were used for subsequent chemical analysis.

Experimental design and statistical analysis

RSM is a collection of mathematical and statistical techniques to utilize quantitative information from an appropriate experimental design to identify optimum conditions. The influence of temperature (60-80 ºC), extraction time (60-100 min), LSR (20-40 %,v/w), and pH (2-3) were determined through an RSM, and Central Composite Design (CCD), requiring a total of 30

experimental runs employed to determine the best combination of parameters for the extraction process. The responses and the process variables are optimized using analysis of variance (ANOVA) to predict the statistical parameters. The independent variable ranges were selected based on Fakayode and Abobi (2018). CCD involves six factorial points, eight axial points, and six points at the center were carried out with the alpha factor of 1.414. All factors have to be adjusted at five coded levels (-α, -1, 0, +1, + α) (*Nahar et al.,* 2017).

$$N = 2^m + 2m + m_c = 16 + 2 \times 4 + 6 = 30 \qquad (2)$$

Where N is the total number of experiments requeird, m is the number of variables and m_c is the number of replicates. The relationship between the variables and the predicted responses were determined by a second-order polynomial equation.

$$Y = b_0 + b_1 x_1 + b_2 x_2 + b_3 x_3 + b_4 x_4 + \qquad (3)$$
$$b_{11} x_1^2 + b_{22} x_2^2 + b_{33} x_3^2 + b_{44} x_4^2 + b_{12} x_1 x_2 + b_{13} x_1 x_3 +$$
$$b_{14} x_1 x_4 + b_{23} x_2 x_3 + b_{24} x_2 x_4 + b_{34} x_3 x_4$$

Where Y is the predicted response (i.e. Yield and DE), b_0 is the constant coefficient, b_1, b_2, b_3, and b_4 are the linear coefficients, b_{11}, b_{22}, b_{33}, and b_{44} represent the quadratic coefficients, b_{12}, b_{13}, b_{14}, b_{23}, b_{24}, and b_{34} represent the second-order interaction coefficients, x_1, x_2, x_3, and x_4 are independent variables.

The outcomes were summarized and statistically analyzed by using Design Expert version 11 software (Stat-Ease Inc., Minneapolis, USA). The ANOVA test was employed to estimate the statistical significance of the regression model. The coefficient of determination R^2, adjusted R^2, and predicted coefficient R^2, lack of fit from ANOVA were used in the determination of the quality of the developed model.

Artificial Neural Network (ANN) modeling

In the present study, the data generated from the experimental design planned through CCD (Table 3) were used to constitute the optimal architecture of ANN. ANN has been applied for the purpose of simulation on the same experimental data used for RSM. The neural network architectures were trained by Levenberg–Marquardt back-propagation algorithm. The network architecture consisted of an input layer of four neurons (Temperature,

Table 2: The coded and the actual form of the independent variables.

S. No	Code variables	Actual level of variables
1	$-\alpha$	$\left(\dfrac{X_{max} + X_{min}}{2}\right) - \left(\dfrac{X_{max} - X_{min}}{2}\right) \times 2^{\frac{n}{4}}$
2	-1	X_{min}
3	0	$\left(\dfrac{X_{max} + X_{min}}{2}\right)$
4	$+1$	X_{max}
5	α	$\left(\dfrac{X_{max} + X_{min}}{2}\right) + \left(\dfrac{X_{max} - X_{min}}{2}\right) \times 2^{\frac{n}{4}}$

Notes: n is the number of variables for any particular experiment, n = 4

extraction time, LSR, and pH), an output layer of two neurons (pectin yield and DE), and a hidden layer. 60% of data points were selected for training to develop the neural network, 20% of the data set was used for validation, and 20% of data sets were for testing. The number of neurons in the hidden layer can be calculated from the expression below:

$$2 (n + m)^{0.5} \quad \text{to} \quad 2n + 1 \tag{4}$$

Where n is the number of neurons in the input layer and m is the number of neurons in the output layer (*Sundarraj et al.*, 2018a). A network is built each of them is trained separately, therefore, the best network was selected based on the accuracy of the predictions within the testing phase. The correlation coefficient among the dependent and independent parameters may be improved by normalizing the distribution data. The input and target data for the individual ANN nodes were normalized within a range of 0 (new x_{min}) to 1 (new x_{max}) in order to achieve fast convergence to obtain the minimal RMSE values. The following equation ensures uniform attention during the training process.

$$x_{norm} = \frac{x_i - x_{min}}{x_{max} - x_{min}} (new \ x_{max} - new \ x_{min}) + \tag{5}$$

$$new \ x_{min}$$

Where x_{norm} is the normalized data, x_i is the input/output data (data of independent and dependent variables), x_{max} and x_{min} are the maximum and minimum values of the particular variable, respectively.

The normalization of inputs and targets was performed to avoid overflows that may appear due to very large or very small weights. The training process was run until a minimum of the MSE was reached in the validation process. All calculations were done using the Neural Network Toolbox of MAT LAB version 8.1(R2013a) utilized throughout the study (*Joel et al.*, 2018).

Comparative analysis of RSM and ANN models

The error analyses such as RMSE, MAE, R^2, SEP, and AAD were carried out between experimental and predicted data in order to evaluate the goodness of fitting and prediction accuracy of the constructed models. The formulas used for error analyses were calculated by Eqs. (6) to (10) (*Liew et al.*, 2014b). To study the modeling abilities of the RSM and ANN models, the values are plotted against the corresponding experimental values.

$$SEP = \frac{RMSE}{Y_e} \times 100 \tag{6}$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n} (Y_{i,p} - Y_{i,e})^2}{\sum_{i=1}^{n} (Y_{i,p} - Y_e)^2} \tag{7}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n} (Y_{i,e} - Y_{i,p})^2}{n}} \tag{8}$$

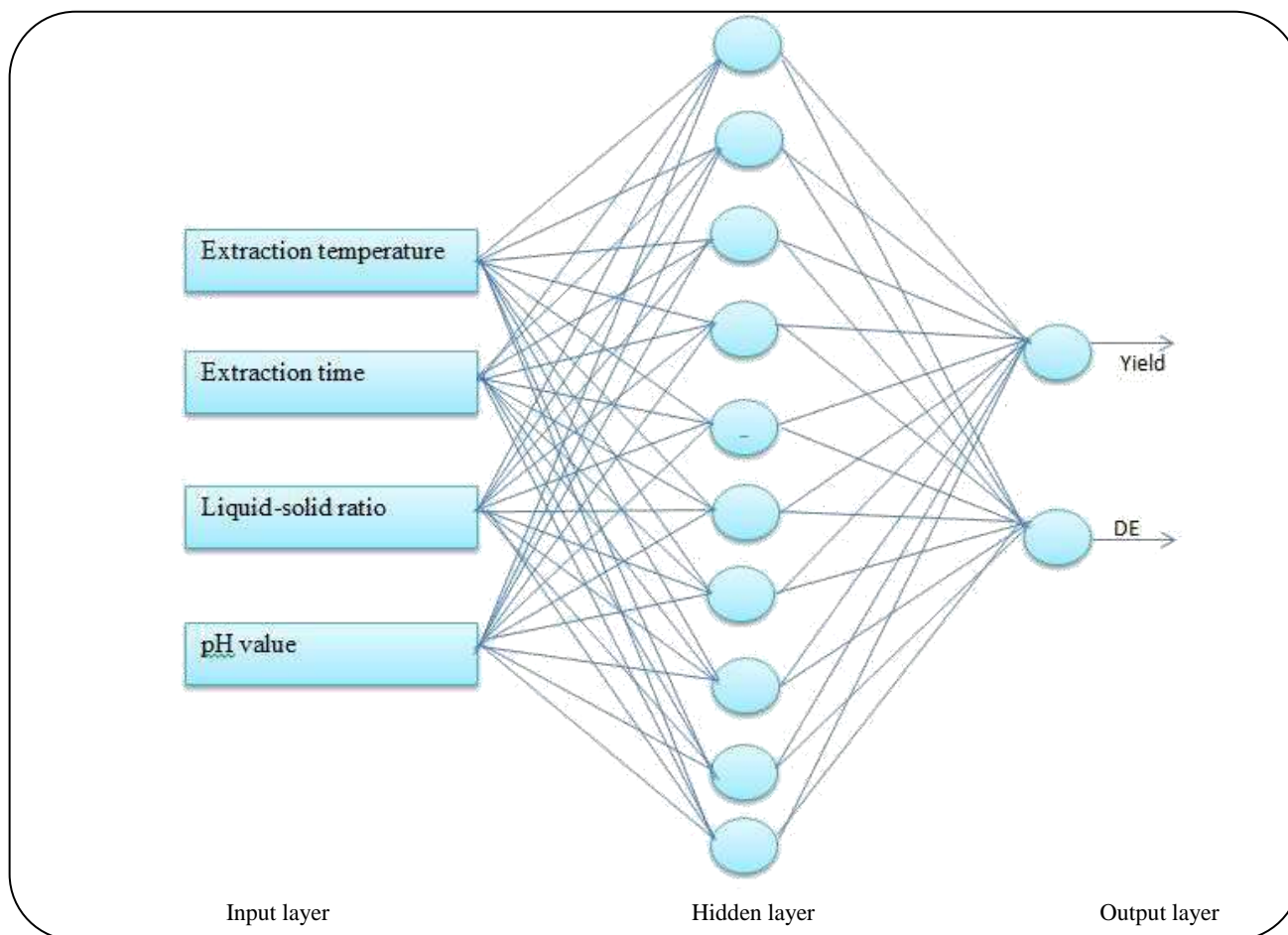$$MAE = \sum_{i=1}^{n} \left(\frac{Y_{i,e} - Y_{i,p}}{n} \right) \tag{9}$$

Fig. 1: Feed-forward with the backward propagation neural network used in the current study.

$$AAD = \frac{100}{n} \sum_{i=1}^{n} \frac{\left| Y_{i,p} - Y_{i,e} \right|}{\left| Y_{i,e} \right|} \qquad (10)$$

where, $Y_{i,e}$ is the experimental data, $Y_{i,p}$ is the predicted data obtained from either RSM or ANN, Y_e is the mean value of experimental data and n is the number of the experimental data. Generally speaking, the smaller the RMSE and the SEP, the higher the modeling ability of a given model. The final network was selected based on the lowest error in the train and depending upon the test data.

RESULTS AND DISCUSSION
Pectin yield

The yields of pectin extracted and DE using microwave-assisted methods from banana peel powder ranges from 5.85 to 16.25% and 39.06 to 66.76, respectively, of the dry weight of the peel depending on the various extraction conditions. RSM has been widely adopted to investigate the effects of several design factors influencing a response by varying them simultaneously in a limited set of experiments.

RSM modeling fitting

The statistical analyses show that quadratic models fit very well into the data for the response. The smaller the p-value and the higher the value of F, the more significant the corresponding coefficient. The ANOVA results showed a perfect fit of the quadratic regression model for banana peel pectin (*F*-value of 70.68) ($p < 0.0001$) and F-value 57.66 ($p < 0.0001$) yield and DE, respectively. In this study, the p-value of "Lack of Fit" for pectin yield and DE were 2.63 ($p > 0.1491$) and 1.94 ($p > 0.2406$), respectively, indicating that lack of fit was not significant relative to the pure error. Therefore, the results obtained verified that the mentioned models (Eqs. (11) and (12)) were accurate enough to predict the pectin yield and DE within the range of the variables studied. The predicted quadratic model for the two responses was highly

Table 3: Central Composite design matrix and experimental yields.

Run	Coded variable					Decoded Variable				Dependent Variable	
	A	B	C	D	Temp (ºC)	Time (min)	LSR (v/w)	pH	Yield (%)	DE (%)
1	-1	-1	-1	-1	60	60	20	2	7.23	44.86
2	1	-1	-1	-1	80	60	20	2	12.72	46.46
3	-1	1	-1	-1	60	100	20	2	9.33	49.16
4	1	1	-1	-1	80	100	20	2	12.45	48.16
5	-1	-1	1	-1	60	60	40	2	10.45	49.56
6	1	-1	1	-1	80	60	40	2	11.92	48.06
7	-1	1	1	-1	60	100	40	2	13.16	55.36
8	1	1	1	-1	80	100	40	2	12.47	53.86
9	-1	-1	-1	1	60	60	20	3	5.85	61.56
10	1	-1	-1	1	80	60	20	3	10.88	62.66
11	-1	1	-1	1	60	100	20	3	8.76	63.56
12	1	1	-1	1	80	100	20	3	10.13	64.76
13	-1	-1	1	1	60	60	40	3	10.46	59.96
14	1	-1	1	1	80	60	40	3	9.49	65.36
15	-1	1	1	1	60	100	40	3	12.75	65.86
16	1	1	1	1	80	100	40	3	10.15	64.76
17	-α	1	0	0	50	80	30	2.5	12.06	61.56
18	α	1	0	0	90	80	30	2.5	16.25	62.16
19	0	-α	0	0	70	40	30	2.5	6.05	56.96
20	0	α	0	0	70	120	30	2.5	8.55	62.66
21	0	0	-α	0	70	80	10	2.5	8.25	53.96
22	0	0	α	0	70	80	50	2.5	13.15	59.86
23	0	0	0	-α	70	80	30	1.5	9.91	39.06
24	0	0	0	α	70	80	30	3.5	7.63	66.76
25	0	0	0	0	70	80	30	2.5	14.26	59.16
26	0	0	0	0	70	80	30	2.5	14.16	59.26
27	0	0	0	0	70	80	30	2.5	13.85	57.66
28	0	0	0	0	70	80	30	2.5	14.34	57.46
29	0	0	0	0	70	80	30	2.5	13.5	60.12
30	0	0	0	0	70	80	30	2.5	13.89	58.16

Table 4: Analysis of variance for response surface quadratic model of pectin yield and DE.

Source	Sum of Squares	Df	Mean Square	F-value	p-value
(A) Yield					
Model	203.49	14	14.53	70.68	< 0.0001
A-Temperature	17.68	1	17.68	85.98	< 0.0001
B-Extraction time	9.63	1	9.63	46.81	< 0.0001
C-liquid-solid ratio	22.62	1	22.62	110.00	< 0.0001
D-pH	10.43	1	10.43	50.71	< 0.0001
AB	6.03	1	6.03	29.31	< 0.0001
AC	19.80	1	19.80	96.29	< 0.0001
AD	2.69	1	2.69	13.08	0.0025
BC	0.3080	1	0.3080	1.50	0.2399
BD	0.0000	1	0.0000	0.0001	0.9913
CD	0.0576	1	0.0576	0.2801	0.6044
A²	0.2011	1	0.2011	0.9779	0.3384
B²	72.71	1	72.71	353.56	< 0.0001
C²	16.61	1	16.61	80.76	< 0.0001
D²	43.59	1	43.59	211.96	< 0.0001
Residual	3.08	15	0.2056		
Lack of Fit	2.59	10	0.2591	2.63	0.1491
Pure Error	0.4934	5	0.0987		
Cor Total	206.57	29			
(B) DE					
Model	1425.79	14	101.84	57.66	< 0.0001
A-Temperature	1.21	1	1.21	0.6879	0.4199
B-Extraction time	61.44	1	61.44	34.79	< 0.0001
C-liquid-solid ratio	46.48	1	46.48	26.32	0.0001
D-pH	1181.61	1	1181.61	669.00	< 0.0001
AB	5.06	1	5.06	2.87	0.1111
AC	0.1600	1	0.1600	0.0906	0.7676
AD	5.06	1	5.06	2.87	0.1111
BC	2.89	1	2.89	1.64	0.2203
BD	4.20	1	4.20	2.38	0.1438
CD	13.69	1	13.69	7.75	0.0139
A²	9.12	1	9.12	5.16	0.0382
B²	0.1129	1	0.1129	0.0639	0.8038
C²	11.98	1	11.98	6.78	0.0199
D²	75.66	1	75.66	42.84	< 0.0001
Residual	26.49	15	1.77		
Lack of Fit	21.06	10	2.11	1.94	0.2406
Pure Error	5.43	5	1.09		
Cor Total	1452.28	29			

Table 5: Regression coefficients of the predicted second-order model for the response variables.

S. No	Response parameter	Pectin yield	Degree of esterification
1	Std. Dev.	0.4535	1.33
2	Mean	11.14	57.29
3	C.V%	4.07	2.32
4	R^2	0.9851	0.9818
5	Adjusted R^2	0.9711	0.9647
6	Predicted R^2	0.9243	0.9111
7	Adeq Precision	31.1883	29.9196
8	Model suggested	Quadratic	Quadratic

significant ($p < 0.0001$). The analysis shows that for pectin yield, A, B, C, D, AB, AC, AD, B^2, C^2, D^2 were found to have a significant effect on the pectin yield while BC, BD, CD, and A^2 were no significant influence on the pectin yield. In the case of degrees of esterification B, C, D, CD, A^2, C^2, D^2 were found to have a significant effect on the DE, while A, AB, AC, AD, BC, BD, and B^2 were not significant (Table 4).

Table 5 shows that the coefficient of variation (CV %) and standard deviation for the two responses in this study was reasonably low and acceptable, indicating better precision and reliability of the experiment. The regression model was found to be highly significant with the R^2 value of pectin yield and DE being 0.9851, and 0.9818, respectively, indicating a close agreement between the observed and the theoretical values predicted by the model equation. Moreover, the value of the adjusted R^2 for pectin yield and DE was 0.9711, and 0.9647 respectively, which confirmed that the model was highly significant, indicating good agreement between the experimental and predicted values of the dependent variables.

Adjusted R^2 and predicted R^2 should be within 20% to be in good agreement as suggested by (*Owolabi et al.,* 2018). This requirement is satisfied in this study with a predicted R^2 value of pectin yield and DE was 0.9243, and 0.9111 respectively.

Development of regression model equation

The experimental results obtained from the pectin yield and DE based on CCD is presented in Table 3. The second-order polynomial function representing pectin yield (Y), DE can be expressed as a function of the four independent variables, namely temperature (A), extraction time (B),

LSR (C), and pH (D) in terms of coded factors after excluding the insignificant terms were given in Eqs. (11) and (12), respectively. It should be noted that Eqs. (11) and (12) are only valid within the range of tested conditions: 50ºC < temperature < 90ºC, 40 min < extraction time < 120 min, 10 < liquid–solid ratio< 50, and 1.5 < pH < 3.5.

$$\text{Pectin yield} = +14.00 + 0.8583A + 0.6333B + \quad (11)$$
$$0.9708C - 0.6592D - 0.6137AB - 1.11AC - 0.410AD - 1.63B^2 - 0.7781C^2 - 1.26\ D^2$$

$$\text{Degree of Esterification} = +58.64 + 1.60B + 1.39C + \quad (12)$$
$$7.02D - 0.9250CD + 0.5767A^2 - 0.6608C^2 - 1.66D^2$$

The positive signs in the models signify the synergetic effects of the factor, while the negative sign indicates the antagonistic effect.

The Figs. 2 and 3 outcomes demonstrated that the predicted values were in very good agreement with the experimental values, in which all the data points are concentrated near the diagonal line, and no scattered points were observed. The points of all predicted and actual responses fell in 45º lines, indicating that the developed model is appropriate to predict the pectin yield and DE. From the graph, it is clear that the values derived experimentally match closely with that developed by the model. Similar studies have been reported for pectin extraction from jackfruit waste (*Sundarraj et al.,* 2018) and pomelo peels (*Nikolova* and *Georgieva*, 2014).

Response surface analysis of the extraction process

The 3D response surfaces plots help to understand the linear, quadratic, and interaction effects on the responses in a straightforward way. The regression models
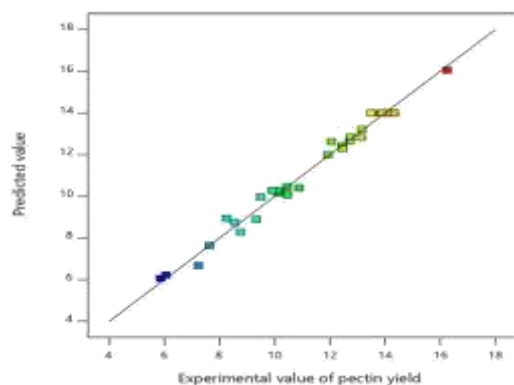
Fig. 2: Correlation between the experimental and predicted value of pectin yield.

Fig. 3: Correlation between the experimental and predicted value of DE.

developed in this study have four independent variables; two variables are held constant at the optimum level, whereas the other two factors are varied within their experimental ranges.

Effect of extraction condition on the pectin yield

The pectin yield obtained in this experiment was found to be in the range of 5.85-16.25% (Table 3), which is comparable to the ripe mango peel pectin (10.76-30.43%) (*Nahar et al.,* 2017), *Citrus sinensis* peels (12.93–29.05%) (*Fakayode* and *Abobi*, 2018) and mango peel pectin (6.1-16.3%) (*Girma* and *Worku*, 2016; *Sangheetha et al.*, 2018). This study is in agreement with previous studies (*Sangheetha et al.,* 2018). Pectin extracted from banana peel was lower than *Azanza Garckeana* (24.38 and 26.75%) *(Joel et al.,* 2018), grapefruit peels (25%) (*Mohamed*, 2016), and Ubá mango peel (18.8 - 32.1%) at different cooking conditions (*Liew et al.,* 2014b), but higher than that of *Durio zibethinus* (2.27-9.35%,w/w) (*Wai et al.,* 2009) and passion fruit peels (7.12-7.16%) (*Wai et al.,* 2009). According to *Happi Emaga et al.* (2008) reported that the yield of pectin extracted from the banana peel ranges from 2.4-21.7% while *Khamsucharit et al.* (2018) reported that the yield of banana peel pectin ranged from 15.89 to 24.08%. These differences may be due to the nature of the fruits and extraction processing conditions.

According to the results presented herein, it is evident that the pectin yield of banana peel was comparable to values obtained from the conventional sources of pectin (i.e. apple pomace, sugar beet, and citrus peel) thus, signifying the po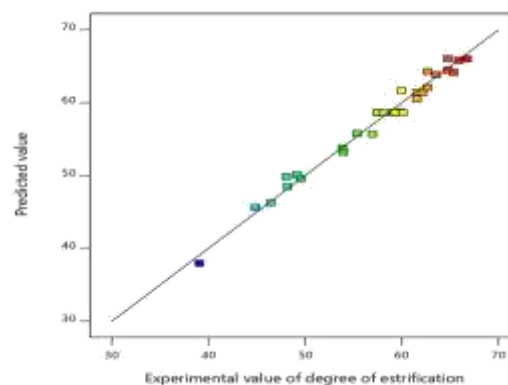tential use of banana peel as an alternative source for the commercial-scale pectin production. Pectin yield was directly proportional to temperature extraction time and liquid–solid ratio and indicated that increasing any of those parameters would result in an increase in the pectin yield until the optimum value was achieved (Eq.11). The yields always increased if temperature, liquid –solid ratio, and extraction time increased (with the other remaining constant), because each of these factors increases the solubility of the extracted pectin, giving a higher rate of extraction. However, further increase in temperature, liquid –solid ratio, and extraction time decreasing the tendency of pectin yield, since too high extraction time and temperature would lead to the breaking down of pectin molecules as pectin is composed of α-(1-4) linked units of galacturonic acid or methyl ester resulting in pectin of lower molecular size which is not perceptible with alcohol. At lower temperatures, the lower viscosity of pectin might cause poor diffusion between the phases which will lead to a slower rate of extraction.

The result shows that the yield increases with an increase in extraction time as the prospecting naturally present in cells takes time to solubilize and go into the solution. The temperature, liquid–solid ratio, extraction time, and pH show a significant ($p < 0.0001$) effect on the pectin yield. The liquid–solid ratio has the most significant effect on the pectin yield whose F value is 110.00, followed by extraction temperature, pH, and time (Table 4). The liquid–solid ratio was directly proportional to pectin yield and indicated that increasing the value of liquid– solid ratio would result in an increasing percentage of pectin yield (Eq. (11)). A similar effect was noted in the extraction
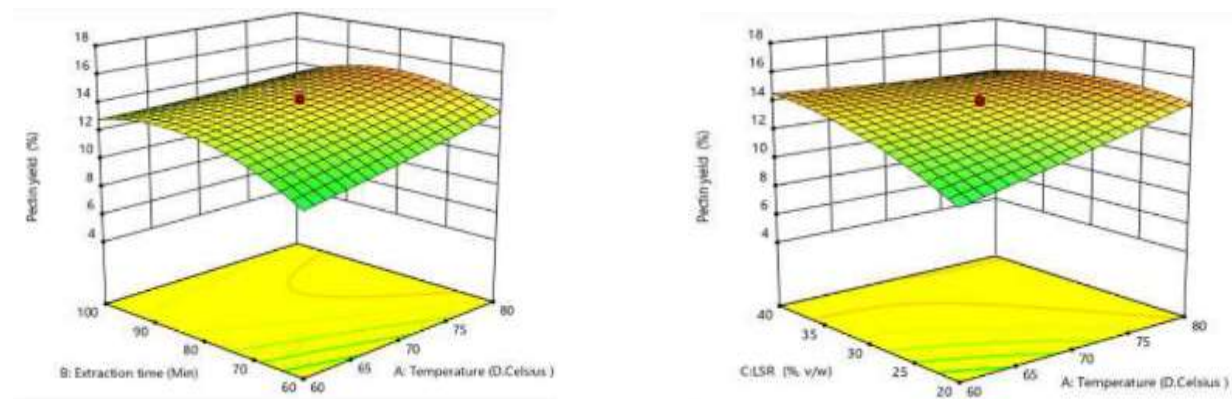
Fig. 4: Three-dimensional response plots for yield as a function of temperature and time at constant LSR and pH.

of pectin from carrot pomace (*Jafari, Khodaiyan, Kiani, & Hosseini*, 2017). The pectin yield decreases with increasing pH value; this is might be due to some pectin that might still be attached to the cell wall components although, pectin molecules can be partially solubilized from plant tissues without degradation in a weak acid solution.

The interaction effects between temperature and liquid –solid ratio, temperature and time, and temperature and pH have a significant ($p < 0.0025$) effect on the pectin yield (Table 4). The interaction between temperature and LSR has the most significant effect on the pectin yield (Table 4). Fig. 4 shows a 3D response surface plot of the pectin yield as a function of temperature and LSR at a fixed extraction time and pH. Increasing the combined effect between temperature and LSR generally decreased the pectin yield; the highest yield was achieved when both variables were at the minimum point. A relatively long period of temperature and LSR would cause a thermal degradation effect on the extracted pectin, thus causing a decrease in the amount perceptible by alcohol. The effect of temperature, pH, and period in this study is similar to the previous work of (*Oliveira et al*., 2015).

The effect of process variable on the degree of esterification

The DE obtained in the experiment is found to be in the range of 39.06 – 66.76 (Table 3). Based on the DE pectin can be classified as LMP with ≤ 50% and HMP with > 50%. The presence of HMP (DE > 50 %) in the extracted banana peel pectin was evident (Table 3). These results were consistent with previous work of 76.30 % DE

in citrus maxima and 79.51%DE in premature lemon pomace pectin (*Azad*, 2014), indicating that banana peel pectins have been classified as HMP similar to those from the citrus peel (62.83%) and apple pomace (58.44%) (*Khamsucharit et al*., 2018).

Based on the ANOVA, the DE of pectin was significantly affected by linear, interactions, and quadratic between process variables. Extraction time, LSR, and pH exhibited significant ($p < 0.0001$) effects on the DE of banana peel pectin (Eq. 12). The DE was positively influenced by extraction time, LSR, and pH. The results obtained from the ANOVA showed that pH has the most significant effect on the DE, followed by extraction time and LSR.

The interaction between LSR and pH exhibited a strong significant ($p < 0.0139$) effect on the DE of pectin. Significant interaction indicates that the factors work independently, whilst the presence of interaction indicates that the difference in DE at different levels of a factor is not the same at all levels of another factor.

The 3D response surface model obtained reflects a linear correlation between the DE with that pH and LSR. This result is in agreement with the observations made by other authors (*Wai et al.*, 2010). Fig. 5 shows a 3D response surface plot of the DE of pectin as a function of LSR and pH at fixed extraction time and temperature. DE was generally decreased as the interaction between LSR and pH increased (Eq. (12)). The quadratic of temperature, LSR, and pH has a significant effect on the DE of pectin. Similar findings were reported by other researchers for banana peel (*Happi Emaga et al.,* 2008) and durian rind pectin (*Wai et al.,* 2010).
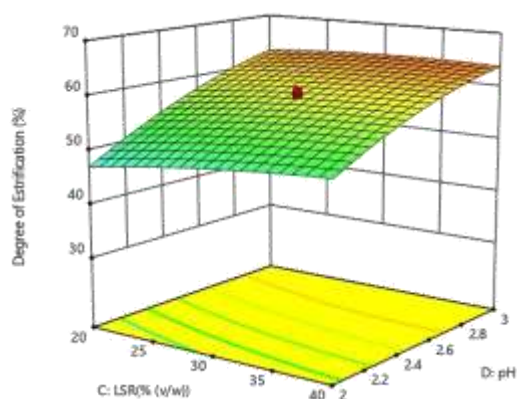
Fig. 5: Three-dimensional response plots for the degree of esterification as a function of LSR and pH at constant extraction time and temperature.

Artificial neural network-based modeling

Fig. 6 shows the spread plot of the experimental versus the computed ANN data in both training, testing, and validation networks. The correlation coefficients (R) values for training (0.99823), validation (0.99851), test (0.99937), and all prediction sets (0.99837) indicate that the ANN model shows better regression and fitting compared to RSM model. Nearly each and every data point have been scattered around the 45° line indicating remarkable compatibility between the experimental and predicted output data values by ANN. Therefore, the ANN prediction for training, validation, and testing is highly substantial and meritorious in terms of correlation and implies that the predicted model was more precise in predicting the responses.

The linear regression analysis between the values predicted by ANN and RSM showed that the values predicted by the ANN model were much closer to experimentally measured data, suggesting that the ANN model has better modeling ability for both simulation and predicted values.

Comparative evaluation of ANN and RSM models

The predictive competence of the ANN and RSM models was determined and compared based on prediction accuracy and various parameters such as RMSE, R^2, SEP, MAE, and AAD. The result showed that both models performed reasonably well, but ANN models have superior modeling capability compared to the RSM models for both pectin yield and DE. As can be observed, the ANN predicted value is much closer to that of the experimentally measured data, suggesting that the ANN model has superior prediction ability than the RSM model (Table 6).

Validation of the optimized condition by response surface modeling

The main objectives of this study were to determine the optimal operating parameters for the maximum pectin yield and DE from banana peel using microwave-assisted extraction. The numerical optimization of the extraction of pectin was performed by using Design Expert 11.0 statistical package by setting the desired goal for each process variable and response. Pectin yield and DE were set at maximum values while the value of process variables were set in the range under study. To validate the statistical experimental strategies, the duplicate was performed under the predicted process conditions.

The maximum predicted pectin yield and DE were achieved at a temperature, extraction time, LSR, and pH of 60°C, 93.8 min, 40% v/w, and 2.72, respectively. Pectin yield and DE were 14.34% (w/w) and 63.58, respectively. The validity of the estimation models built through the statistical experimental design was verified by the small differences ($< 4\%$) between the experimental and the predicted responses. The result indicates that there was good agreement between the predicted and experimental results verifying the validity of the model. Compared to the data obtained from the literature, the optimum extraction conditions of the pectin in the accepted limits of banana peels (Musa AAA) as reported by *Happi Emaga et al.* (2008) and *Oliveira et al.* (2015)

Physicochemical characterization of banana peel pectin

The physiochemical characterization of pectin was carried out at optimized operating conditions. The moisture content of pectin extracted in this experiment was found to be 7.87%, which is slightly higher than banana peels of different varieties (4.54 – 6.24%) and apple pomace (4.54%) but slightly lower than citrus peel (7.92%) (*Khamsucharit et al.*, 2018). While moisture content is certainly important in the safe storage and quality of pectin, the actual relationship between equilibrium water activity and equilibrium moisture content is the critical determinant of safe storage and quality (*Mohamadzadeh et al.,* 2010).

Table 6: Comparison of predictive abilities of RSM and ANN models.

Parameters	Pectin yield (%)			Degree of esterification	
	RSM	ANN		RSM	ANN
RMSE	0.1473	0.1048		0.1527	0.1105
R^2	0.9851	0.9918		0.9818	0.9901
AAD (%)	1.001	0.8998		0.1899	0.1109
MAE	0.1320	0.1020		0.1215	0.0770
SEP	1.097	0.8105		0.2387	0.1864
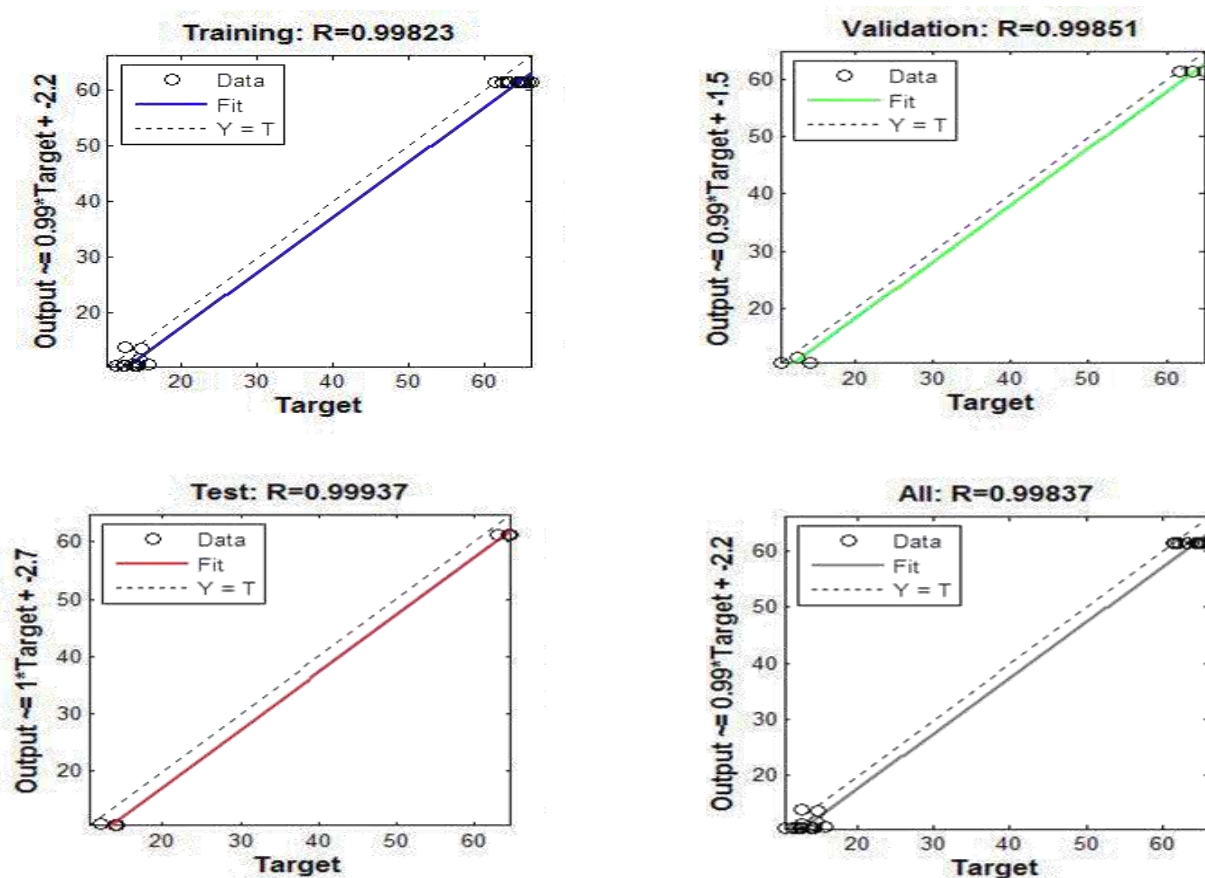
*Fig. 6: Neural Network model with training, validation, test, and all prediction sets.*

The ash content of pectin extracted from banana peel was found to be 1.44% (Table 8) which was in a similar range to that obtained from the conventional pectin sources, apple pomace (1.96%) and citrus peel (3.46%). The current finding was in agreement with an earlier finding of various banana peel pectin (1.43-2.76%) (*Khamsucharit et al.,* 2018; *Phaiphan et al.,* 2019). Low ash content (below 10%) was more favorable for gel formation, it could be reduced by washing with acidified alcohol (*Manh et al.,* 2019).

The Anhydrouronic acid content (AUA) of pectin extracted from banana peel was found to be 67.43% (Table 8), which is comparable to pectin extracted from banana peels of different varieties (34.56– 66.67%) while lower than citrus peel and apple pomace (*Khamsucharit et al.,* 2018). The AUA indicates the purity of the extracted

Table 8: Physicochemical characterization of pectin extracted from different varieties of banana peel.

Parameters	Current value (%)	Previous value (%) and references		Commercial citrus pectin
Moisture (%)	7.87	4.54 – 6.24%	(*Khamsucharit et al.,* 2018)	7.31±0.73
Ash (%)	1.44	3.39-5.40	(*Phaiphan et al.,* 2019)	1.84±0.05
DE (%)	63.58	51.79 - 72.03	(*Oliveira et al.,* 2015)	78.92±2.16
AUA (%)	67.43	34.56 - 66.67	(*Khamsucharit et al.,* 2018)	76.8 - 82.05
MeO (%)	8.52	3.86 – 14.5	(*Phaiphan et al.,* 2019)	9.06 - 12.88
EW (g/ml)	956.49	943.14 - 1456.93	(*Oliveira et al.,* 2015)	551.29 - 577.72
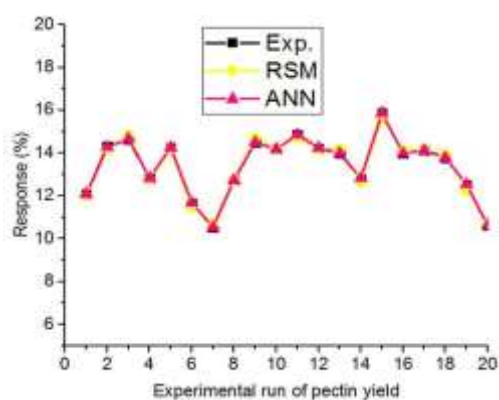
Fig. 7: Comparison of experimental with the predicted value obtained by the RSM and ANN model for the prediction of pectin yield.
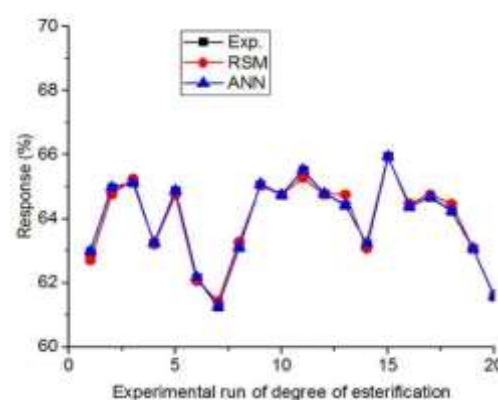
Fig. 8: Comparison of experimental with the predicted value obtained by the RSM and ANN model for the prediction of the degree of esterification.

pectin with a recommended value of not less than 65% for pectin used as food additives or for pharmaceutical purposes (May, 1990). According to Food and Agriculture Organization (FAO), Food Chemical Codex (FCC), and European Union (EU), pectin must contain at least 65% of AUA. In this study, the highest AUA content of banana peel pectin was obtained which lies in the acceptable limits of pectin purity. The extracted pectin from banana peel had higher than 65% and met the criteria for commercial pectin; thus, banana peel can be an alternative source of high methoxyl pectin.

Methoxyl content is an important factor in controlling the setting time of pectin and the ability of the pectin to form gels (Constenla and Lozano, 2003). The methoxyl content of pectin extracted from banana peel was found to be 8.52% (Table 8), which is comparable to pectin extracted from pomelo peel (8.57%), passion (8.81%-9.61%) (*Azad*, 2014), banana peels of different varieties (3.86– 8.46%) while lower than citrus peel (9.06%) and higher than apple pomace (7.92%) (*Khamsucharit et al.,*

2018). Based on methoxyl content value in this study indicates that banana peel pectin was categorized as HMP.

The equivalent weight (EW) of pectin extracted from banana peel was found to be 956.49 which was higher than citrus peel (577) and apple pomace (551) but comparable to other varieties of banana peel pectin (943-1456) (*Khamsucharit et al.,* 2018; *Oliveira et al.,* 2015) and lemon pomace peel pectin (368 -1632) (*Azad*, 2014).

CONCLUSIONS

In this study, the modeling, predictive, and generalization capabilities of RSM and ANN models were compared for microwave-assisted extraction of pectin from banana peel. The performance of both models was compared based on the prediction accuracy of the pectin yield and degree of esterification. The study revealed that all four variables linearly affect the pectin yield and DE significantly compared to the combined and squared effects. Based on the values of R^2, RMSE, SEP, MAE, AAD for validation data sets, ANN model was demonstrated to be more efficient than RSM model

both in data fitting and prediction capabilities. The optimum conditions of the pectin yield and DE were achieved at temperature, extraction time, LSR, and pH of 60ºC, 93.77min, 40% (v/w), and 2.7, respectively, with the desirability of 0.853. Under these conditions, the maximum pectin yield and DE were 14.34% and 63.58, respectively. Based on the value of methoxyl content and degree of esterification the extracted pectin was categorized as high methoxyl pectin. The value of AUA content indicates that the extracted pectin from banana peel had high purity which met the criteria for use as a food and pharmaceutical additive and suggested its potential use as an alternative source of commercial pectin production.

Acknowledgments

REFERENCES

[1] Azad A.K.M., Isolation and Characterization of Pectin Extracted from Lemon Pomace during Ripening. *Journal of Food and Nutrition Sciences*, **2(2)**: 30 (2014). https://doi.org/10.11648/j.jfns.20140202.12

[2] Constenla D., Lozano J.E., Kinetic Model of Pectin Demethylation, *Latin American Applied Research*, **33(2)**: 91–95(2003).

[3] Fakayode O.A., Abobi K E. Optimization of Oil and Pectin Extraction from Orange (Citrus Sinensis) Peels: A Response Surface Approach, *Journal of Analytical Science and Technology*, **9(1)**: (2018). https://doi.org/10. 1186/s40543-018-0151-3

[4] Girma E., Worku T., Extraction and Characterization of Pectin from Selected Fruit Peel Waste, **6(2)**: 447–454 (2016).

[5] González-Montelongo R., Gloria Lobo M., González M., Antioxidant Activity in Banana Peel Extracts: Testing Extraction Conditions and Related Bioactive Compounds, *Food Chemistry*, **119(3)**: 1030–1039 (2010). https://doi.org/10.1016/j.foodchem.2009.08.012

[6] Happi Emaga T., Ronkart S.N., Robert C., Wathelet B., Paquot M., Characterisation of Pectins Extracted from Banana Peels (Musa AAA) under Different Conditions Using an Experimental Design, *Food Chemistry*, **108(2)**: 463–471 (2008). https://doi.org/10.1016/j.foodchem.2007.10.078

[7] Jafari F., Khodaiyan F., Kiani H., Hosseini S.S., Pectin from Carrot Pomace: Optimization of Extraction and Physicochemical Properties, *Carbohydrate Polymers*, **157**: 1315–1322 (2017). https://doi.org/ 10.1016 /j.carbpol.2016.11.013

[8] Joel J.M., Barminas J.T., Riki E.Y., Yelwa J.M., Edeh F., Extraction and Characterization of Hydrocolloid Pectin from Goron Tula (Azanza garckeana) Fruit. *World Scientific News*, *101*(June), 157–171 (2018). Retrieved from www.worldscientificnews.com

[9] Khamsucharit P., Laohaphatanalert K., Gavinlertvatana P., Sriroth K., Sangseethong K., Characterization of Pectin Extracted from Banana Peels of Different Varieties, *Food Science and Biotechnology*, **27(3)**: 623–629 (2018). https://doi.org/10.1007/s10068-017-0302-0

[10] Liew S.Q., Chin N.L., Yusof Y.A., Extraction and Characterization of Pectin from Passion Fruit Peels, *Italian Oral Surgery*, **2**: 231–236 (2014a). https://doi.org/10.1016/j.aaspro.2014.11.033

[11] Liew S.Q., Chin N.L., Yusof Y.A., Extraction and Characterization of Pectin from Passion Fruit Peels, *Agriculture and Agricultural Science Procedia*, **2**: 231–236 (2014b). https://doi.org/10.1016/j.aaspro.2014 .11.033

[12] Manh B., Nguyen N., Pirak T., Cogent Food & Agriculture Physicochemical Properties and Antioxidant Activities of White Dragon Fruit Peel Pectin Extracted with Conventional and Ultrasound-Assisted Extraction Physicochemical Properties and Antioxidant Activities of White Dragon Fruit P, *Cogent Food & Agriculture*, **5(00)**, (2019). https://doi.org/10.1080/23311932.2019.1633076

[13] May C.D., Industrial Pectins: Sources, Production and Applications, Carbohydrate Polymers. *J. Ind. Appl. Pectin*, **12(1)**, 79–99 (1990).

[14] Mohamadzadeh J., Sadeghi-Mahoonak A.R., Yaghbani M., Aalami M., Extraction of Pectin from Sunflower Head Residues of Selected Iranian Cultivars, *World Applied Science Journal*, **8(1)**: 21–24 (2010).

[15] Mohamed H., Extraction and Characterization of Pectin from Grapefruit Peels, *MOJ Food Processing & Technology*, **2(1)**: 31–38 (2016). https://doi.org/10.15406/mojfpt. 2016.02.00029

[16] Mohamed Hasan Z., Extraction and Characterization of Pectin from Various Tropical Agrowastes, *ASEAN Food Journal*, **2**: 143–150 (1995).

[17] Nahar K., Haque M., Nada K., Uddin M., Mansur M.A. Al, Khatun N., Jabin S., Pectin from Ripe Peels of Mango Cultivars, *Bangladesh Journal of Scientific and Industrial Research*, **52(3)**: 229–238 (2017). https://doi.org/10.3329/bjsir.v52i3.34159

[18] Nikolova I., Georgieva N., Optimization of the Pectin Extraction from Pomelo Peels by Oxalic Acid and Microwave, *Banat?S Journal of Biotechnology*, **5(9)**: 67–73 (2014). https://doi.org/10.7904/2068

[19] Oliveira T.Í.S., Rosa M.F., Cavalcante F.L., Pereira P.H.F., Moates G.K., Wellner N., Mozzetto S.E., Waldron K.W., Azeredo H.M.C.,Azeredo H.M.C., Optimization of Pectin Extraction from Banana Peels with Citric Acid by Using Response Surface Methodology, *Food Chemistry*, **198**: 113–118 (2015). https://doi.org/10.1016/j.foodchem.2015.08.080

[20] Owolabi R.U., Usman M.A., Kehinde A.J., Modelling and Optimization of Process Variables for the Solution Polymerization of Styrene Using Response Surface Methodology, *Journal of King Saud University - Engineering Sciences*, **30(1)**: 22–30 (2018). https://doi.org/10.1016/j.jksues.2015.12.005

[21] Phaiphan A., Churat S., Doungta T., Wichalin P., Khanchai W., Penjumras P., Effects of Microwave and Ultrasound on the Extraction of Pectin and its Chemical Characterisation of Banana (Musa Sapientum L.) Peels, *Food Research*, **4(6)**: 2030–2036 (2019). https://doi.org/10.26656/fr.2017.4(6).248

[22] Control for Fruits and Vegetable Products (2nd ed.). In: *ASEAN Food Journal*.

[23] Ranganna S., *Handbook of Analysis and Quality Control for Fruit and Vegetable Products, Second Edition* (2004).

[24] Sangheetha S., Illeperuma D.C.K., Navaratne A.N., Jayasinghe C., Effect of pH, Temperature and Time Combinations on Yield and Degree of Esterification of Mango Peel Pectin: A Box-Behnken Design Based Statistical Modelling, *Tropical Agricultural Research*, **30(2)**: 1 (2018). https://doi.org/10.4038/tar.v30i2.8304

[25] Sundarra, A.A., Thottiam Vasudevan R., Sriramulu G., Optimized Extraction and Characterization of Pectin from Jackfruit (Artocarpus Integer) Wastes Using Response Surface Methodology, *International Journal of Biological Macromolecules*, **106**: 698–703 (2018). https://doi.org/10.1016/j.ijbiomac.2017.08.065

[26] Vriesmann L.C., Teófilo R.F., Lúcia de Oliveira Petkowicz C., Extraction and Characterization of Pectin from Cacao Pod Husks (Theobroma cacao L.) with Citric Acid, *LWT - Food Science and Technology*, **49(1)**: 108–116 (2012). https://doi.org/10.1016/j.lwt.2012.04.018

[27] Waghmare A.G., Arya S.S., Utilization of Unripe Banana Peel Waste as Feedstock for Ethanol Production, *Bioethanol*, **2(1)**: 146-156 (2016). https://doi.org/10.1515/bioeth-2016-0011

[28] Wai W. W., Alkarkhi A.F.M., Easa A.M., Optimization of Pectin Extraction from Durian Rind (Durio Zibethinus) Using Response Surface Methodology, *Journal of Food Science*, **74(8)**: 637–641 (2009). https://doi.org/ 10.1111/j.1750-3841.2009.01331.x

[29] Wai W.W., Alkarkhi A.F.M., Easa A.M., Effect of Extraction Conditions on Yield and Degree of Esterification of Durian Rind Pectin: An Experimental Design, *Food and Bioproducts Processing*, **88(2–3)**: 209–214 (2010). https://doi.org/10.1016/j.fbp.2010.01.010

[30] Woldu Z., Mohammed A., Belew D., Shumeta Z., Assessment of Banana Production and Marketing in. *International Journal of Sciences: Basic and Applied Research*, **4531**, 283–307 (2015). htpp//gasser.org/index.php? journal