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ULTRASOUND ASSISTED EXTRACTION OF PECTIN FROM DRAGON FRUIT PEELS

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Abstract

Pectin has increasingly gained importance as it is largely employed in numerous industrial applications due to its gelling capability. Existing literature and studies have shown that dragon fruit peel is an ideal source of pectin. However, there is a lack of studies on the extraction of pectin by ultrasound assisted extraction (UAE) method. The main focus of this research was to study the effect of various parameters on the extracted pectin yield. At the same time, the process was optimised through the statistical approach of response surface methodology (RSM). A three-factor five-level central composite design (CCD) was employed to study the effects of temperature (60-80°C), extraction time (10-30 min) and liquid-solid ratio (15:1-45:1 mL/g). Analysis of variance (ANOVA) was performed to study the significance of independent variables on pectin yield. Pectin yield was significantly affected by extraction temperature and time but not by liquid-solid ratio (LSR). The optimised condition was identified to be an extraction temperature of 71.8°C with time and LSR of 25 min and 35.6:1 mL/g, respectively. Optimum yield of 7.49% was retrieved at the said condition. Further evaluation of the optimised condition was validated by ANOVA. The kinetics was also investigated through the application of five commonly used extraction models (First-order kinetic model, Peleg's model, Second-order rate equation, Logarithmic model and Power law equation). Validation of the kinetic models was based on determination of coefficient (R^2). Peleg's model with highest R^2 (0.9642) was determined to be the most suitable model to describe the extraction process of pectin from studied fruit peels. Present study demonstrated that dragon fruit peel is an ideal alternative source of pectin. An efficient UAE process had been established to extract pectin that can bring contribution to the future pectin industry.

Keywords: Central composite design, Pectin, dragon fruit peels, ultrasound assisted extraction.

1. Introduction

Pectin, naturally occurring polysaccharide, is a treasured by-product that can be obtained from fruit wastes [1]. It is a complex polysaccharide that consists of a a-1,4 linked D-galacturonic acid units backbone which is interrupted by α-1-2-L- rhamnose units that is attached to the side chain of a neutral sugar [2]. In addition, pectin can be esterified with methyl ester at carbon six or acetylated at carbon two or three positions [2]. The name, pectin comes from the Greek word 'pektos' that is defined as hard and firm, indicating the capability of pectin to form gel [3].

Recently pectin has increasingly gained importance as it is largely employed in food and pharmaceutical industry due to its gelling capability [4]. In 2011, around 60% of the pectin produced was used in food industry as gelling agent. Pectin's best-known use is in jams as it is the only permitted gelling agent in these products. On the other hand, approximately 25% of pectin in the market is used in beverages as thickening agent to give texture to soft drinks or juices. Another 4% is contributed to dairy products as stabiliser by forming electrostatic interaction with casein particles. This prevents the alteration of casein particles and unstable dispersion caused by heat treatment and acidification. Furthermore, the use of pectin in pharmaceutical industry is growing due to its potential drug delivery properties. Recently, pectin is also applied in improving the shelf life of vegetables and fruits as it can reduce oxygen and water transfer by forming a barrier [3].

The major source of pectin in current market is citrus peels and apple pomace which are by-products obtained from the manufacturing of juice or cider [2, 5]. High pectin content in both fruit peels makes them an important source of pectin. Apple pomace contains 10-15% of pectin on a dry matter basis while citrus peel contains 20-30% [6]. Due to the high demand of pectin in health and pharmaceutical industry, more investigations are carried out to evaluate pectin content from other fruit sources such as lychee, mango, papaya, banana (*Musa sp.*) and others [2].

Dragon fruit (*Hylocereus polyrhizus*) is widely grown in Asian countries such as Malaysia, Vietnam, Thailand, the Philippines and Taiwan [7]. In Malaysia, the average production of dragon fruit in 2014 was about 3,835 tons [8]. Besides, the peel constitutes about a quarter of the fruit mass [9]. The peels of dragon fruit are waste from fruit processing industry, which in turn leads to an environmental problem. Thus, pectin extraction from the peels of dragon fruit can add value and minimise the waste disposal [10]. Additionally, dragon fruit is chosen in this study due to the potential of becoming important source of pectin in the future. In a study by [10], dragon fruit proved to be a great potential in food industry. The extracted pectin exhibited a high quantity of properties including 67.5% of galaturonic acid (GA) content and 49.84% degree of esterification (DE) which were comparable to citrus pectin (53.62% DE). Besides, another study showed the yield of pectin from dragon fruit peels was 11 - 13% that was equivalent to apple pomace [5]. While, available literature has established the beneficial properties of pectin yield and dragon fruit as its source, there are limited investigations on the effective extraction of the said compound.

There are several types of extraction methods that have been employed such as conventional extraction [11], microwave-assisted extraction (MAE) and ultrasound-assisted extraction (UAE) [12]. From extensive literature, it was found that only a few fruits have been studied to extract pectin using UAE such as grapefruit [13], yellow passion fruit [14], grape pomace [15], pomegranate fruit [16], fresh grapefruit

[17], banana peel [18] and apple [19]. To the best of our knowledge, no UAE method has been performed for pectin extraction from dragon fruit peels. UAE is a less expensive method where acoustic energy is subjected to solvent to perform pectin extraction from sample [20]. Ultrasound can assist in the penetration of solvent into sample through cavitation and extract the solute, thereby, increasing the extraction yield. Furthermore, previous studies showed that the application of UAE resulted in shorter extraction time and higher yield when compared to other methods [13, 21]. With regards to this, UAE was performed in this study.

Extraction parameters such as temperature, extraction duration and liquid-solid ratio can affect the yield of pectin. Therefore, the primary aim of current work was the application of response surface methodology (RSM) using central composite design (CCD) to evaluate the variables. RSM is an useful, inexpensive and effective statistical tool for optimization process [22]. Three independent variables (extraction temperature, extraction duration and liquid-solid-ratio) were evaluated with the yield of pectin in order to get the optimum extraction condition. Present study also aims to investigate the extraction kinetic of pectin from studied fruit peels by fitting the experimental data to five kinetic models namely, first-order kinetic model, Peleg's model, second-order rate law, logarithmic model and power law equation to evaluate the applicability. A suitable extraction model that can explain the extraction of pectin (best fit to the experimental data) from studied raw material was determined.

2. Materials and Methods

2.1. Preparation and pre-treatment of dragon fruit peels

2.1.1. Material collection

Raw material of the target waste fruit, red dragon fruit *(Hylocereus polyrhizus)* peels was collected from local fruit juice vendor "Vitamin Factory" in Taylor's Lakeside University, Selangor.

2.1.2 Drying and grinding

The peels were washed with running tap water and distilled water to remove impurities on the surface. Then they were cut into smaller pieces using a knife to improve the air contact in drying oven for faster drying purpose. Drying was carried out at moderate temperature of 50°C in a convective drying oven (Model UFE-800, Memmert, Germany) until constant weight was attained in order to completely remove moisture to prevent spoilage [23]. The dried peels were sent to a grinder followed by sieving through 500 µm sieve mesh to produce dragon fruit peel powder [10, 24]. The sieved dried powder was stored in a seal bag and kept dry in desiccator prior to extraction [25].

2.2. Ultrasound-assisted extraction (UAE) for preliminary study

Preliminary study was done in order to identify the best range of temperature for further optimisation stage. Dried fruit powder (1 g) was first placed in a schott bottle and mixed with 0.1M citric acid solution of pH 2 with the LSR set at 30:1 mL/g. The powder-acid solution was then placed in an ultrasonic bath (Elmasonic Model P120H, Singen, Germany) to carry out the extraction. Using the variables of LSR (30:1 mL/g), extraction was performed at different temperatures of 50, 60

and 70°C at a constant frequency of 37 kHz. Extract samples were retrieved every 5 min for a total duration of 75 min. Each experiment was carried out thrice to ensure the result reliability.

2.3. Pectin precipitation and pectin yield determination

Pectin precipitation followed method described by [5] with modifications. The extracted mixture was filtered using vacuum filtration. The filtrate was precipitated with 95% of ethanol at equal volume (EV=1) for 2 hrs. The mixture was then centrifuged at 6000 rpm for 30 min. The filtrate was discharged whereas the pellet from previous step was collected and rinsed with 95% ethanol three times to remove impurities such as monosaccharide, disaccharides and phenolic compounds. The wet pectin was dried in the oven again at 60°C until constant weight was obtained. Pectin yield (%) was expressed as the ratio of dried pectin mass (W_p) obtained after extraction to the initial mass of dried peel powder (W_t) used for extraction [26]:

$$\text{Yield (\%)} = \frac{W_p}{W_t} \times 100 \quad (1)$$

2.4. Kinetic modelling for preliminary study

Following the extraction, dried weight was weighted using a digital balance (Ohaus PA4102, U.S.A) [27]. Pectin yield was calculated as shown in section 2.3 and a graph of pectin yield (%) against time (minutes) was plotted to show the extraction pattern of pectin from dragon fruit peels. The temperature that produced the highest pectin yield was selected as the central point of CCD.

2.5. Optimisation with response surface methodology (RSM) and experimental validation

RSM was employed to optimise the extraction conditions of pectin from the studied fruit waste. RSM has been previously used in successful optimisation of extraction parameters from natural sources [28]. Hence, a three-factor five-level CCD in Design expert software v. 7.0 (Stat-Ease, Minneapolis, Minnesota, USA) with three independent variables (extraction temperature, extraction time and LSR) was applied. The five experimental levels in CCD were coded as $-\alpha$ (-1.682), -1 (low), 0 (medium), +1 (high), $+\alpha$ (1.682) where 0 implies the central point [29-31]. Meanwhile, three independent variables of extraction temperature, extraction time and LSR were coded as X_1, X_2 and X_3. The extraction temperature that produced the highest yield in previous kinetic study was 70°C. Thus, the temperature range in CCD was set as 60 to 80°C. Meanwhile, the range of other two variables, extraction time and LSR were chosen based on the previous studies. The range of optimum extraction time was 10 to 30 min [16, 17, 21, 23, 26, 32, 33]. On the other hand, the optimum range of LSR was 15:1 to 45:1 mL/g [16, 17, 23, 33, 34]. Table 1 shows the three variables in regards to their actual and coded values.

After inserting the independent variables and the experimental range, CCD generated the combinations of design parameters with their respective responses. Table 2 represents the CCD for the extraction yield of pectin.

A total of combination set of 20 runs was generated from CCD and each run was conducted in triplicates to obtain the average yield. A mathematical model -

second-order (quadratic) polynomial equation that can estimate the relationship between the independent variables and response was built. Optimisation of the extraction parameters was performed using response optimiser to find the optimum values for each of the independent variables that give the optimum pectin yield. After obtaining the optimum condition from the software, experimental validation was carried out thrice.

Table 1. Experimental range and levels of the independent variables.

Independent Variables	Code	Independent Variables Level				
		$-\alpha$ (-1.68)	**-1**	**0**	**+1**	**$+\alpha$ (+1.68)**
Extraction temperature (°C)	X_1	60	64	70	76	80
Extraction time (min)	X_2	10	14	15	26	30
Liquid-solid ratio (LSR)(mL/g)	X_3	15:1	21	30	39	45:1

Table 2. Central composite design (CCD) experimental design for the extraction of pectin yield.

Std	Run	Temperature (X_1)(°C)	Time (X_2)(min)	LSR (X_3)(mL/g)	Response (yield)(%)
1	2	64	14	21	
2	9	76	14	21	
3	6	64	26	21	
4	12	76	26	21	
5	13	64	14	39	
6	8	75	14	39	
7	17	64	26	39	
8	3	76	26	39	
9	1	60	20	30	
10	16	80	20	30	
11	4	70	10	30	
12	10	70	30	30	
13	7	70	20	15	
14	20	70	20	45	
15	5	70	20	30	
16	11	70	20	30	
17	15	70	20	30	
18	18	70	20	30	
19	14	70	20	30	
20	19	70	20	30	

2.6. Analysis of variance (ANOVA)

ANOVA with 95% of confidence level ($\rho < 0.05$) was used to analyse the data from RSM in order to determine the significance for each of the independent variables. The adequacy of the regression model was evaluated with *F*-value, ρ-value and coefficient of determination (R^2) [35-37]. The *F*-value indicates the ratio of mean

square error to the pure error whilst the ρ-value represents the significance of the variables [35]. R^2 is defined as the degree of fit and is used to describe how close the experimental data fitted regression line [37]. In general, the term with larger *F*-value and smaller ρ-value is regarded as more significant [38]. The range of coefficient of determination (R^2) is between 0 - 1 and the larger value is desirable [37]. Additionally, coefficient of determination (R^2) > 0.75 implies aptness of the developed regression model [14].

2.7. Mathematical modeling for solid-liquid extraction (SLE) of pectin from dragon fruit peel

In this study, five existing kinetic extraction models were used to study the kinetic extraction process of pectin from dragon fruit peels. These models are first-order kinetic model, Peleg's model, second-order rate equation, logarithmic model and power law equation.

2.7.1. First-order kinetic model

A study on extraction of polyphenol from jamum seeds by Balyan & Sarkar (2016) demonstrated that first-order kinetic model was the best model to describe the extraction process [39]. The first-order kinetic model was based on Fick's Law and can be used to study the mass transfer rate of extracted solute into liquid phase from the solid material [40]:

$$C_t = C_\infty(1 - e^{-kt}) \tag{2}$$

where C_t is the concentration of solute in bulk liquid phase at time t (mg solute/g of solid material), C_∞ is concentration of solute at equilibrium at the solid-liquid interface (mg solute/ g of solid material), k is the overall volumetric mass transfer coefficient (min^{-1}), and t is the extraction time.

2.7.2. Peleg's model

A well-known kinetic model that depicted the food material sorption isotherms was introduced by Peleg (1988) [41]:

$$C_t = C_0 + \frac{t}{k_1 + k_2 \cdot t} \tag{3}$$

where C_t is the concentration of solute at time t (mg solute/g of solid material), C_0 is the initial concentration of solute in liquid solvent, t is the extraction time, k_1 is the Peleg's rate constant (min g/mg) whereas k_2 is Peleg's capacity constant (min g/mg).

However, C_0 was omitted from the equation due to the initial concentration of solute in liquid solvent was zero. Thus, the modified Peleg's equation can be written as:

$$C_t = \frac{t}{(k_1 + k_2 t)} \tag{4}$$

Peleg's model is extensively used by researches to explain the extraction curves of biological compound from various plants with modification owing to the similarity to the sorption curves shape. In addition, this model was used by [42] to represent the extraction process of total polyphenol from soybean.

2.7.3. Second-order rate equation

This model was applied by researchers to explain the extraction of antioxidant from pomegranate marc and *Piper betle* [12, 43]. The general second-order kinetic model is shown as below:

$$\frac{dC_t}{dt} = k(C_e - C_t)^2 \quad (5)$$

where k is the rate constant (L/g min), C_e is the concentration of solute at equilibrium in liquid phase (g/L) and C_t is the solute concentration in liquid phase at time t (g/L).

The integrated rate law for this model under the boundary conditions $t = 0$ to t and $C_t = 0$ to C_t, the equation is as below:

$$C_t = \frac{C_e^2 kt}{1 + C_e kt} \quad (6)$$

The initial extraction rate when t approaches zero can be presented as:

$$h = kC_e^2 \quad (7)$$

By rearranging equation 6 and 7, the C_t can be expressed as:

$$C_t = \frac{t}{\left(\frac{1}{h}\right) + \left(\frac{t}{C_e}\right)} \quad (8)$$

2.7.4. Logarithmic model

Logarithmic model was applied successfully to describe the extraction process of polyphenol compound from soybean [42]. The model is shown as below:

$$c(t) = a \log t + b \quad (9)$$

where $c(t)$ is the concentration of solute at time t, a and b are the constant of logarithmic model.

2.7.5. Power law equation

Power law is another useful model to depict the solid-liquid extraction process. Successful example of the application of this model was during the extraction of rutin from Chinese scholar-tree flower [44], water soluble compounds and polysaccharides from medicinal fungus [27] and vanilla extraction from vanilla palifolia Andrews [45]. Some general assumptions were made with regards to Power law equation [27]:

- All solid particles are uniform size with spherical shape
- The extractable material are uniformly distributed within the solid
- The extractable material have constant diffusion coefficient
- The solid particles are well dispersed in the liquid extracting solvent

The Power Law equation can be written as:

$$C_t = Bt^n \quad (10)$$

where C_t represents the solute concentration (mg g-1) at time t, t is the extraction time (min), B is the extraction rate constant (g mg-1min-1), t is the extraction time (min) and n is the power-law exponent (n<1).

2.8. Extraction kinetic model fitting

Five kinetic models mentioned in section 3.7 were solved using SOLVER, which was incorporated in Microsoft Excel 2010. The coefficient of determination (R^2) was chosen to determine the applicability of the model to the experimental data and the models were solved for R^2 close to unity. The R^2 formula is shown as below:

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(C_{exp,i} - C_{pre,i})^2}{\sum_{i=1}^{n}(C_{exp,i} - \bar{C})^2} \quad (11)$$

where n represents the number of samples, $C_{exp,i}$ indicates the experimental pectin concentration, $C_{pre,i}$ is the predicted pectin concentration and $\bar{C}$ symbolises the mean value of experimental data.

3. Result and Discussion

3.1. Preliminary study

The result of the preliminary study of pectin yield from dragon fruit peels is shown in Fig. 1. It was found that the extraction temperature of 70°C gave the highest yield of 6.72% compared to 50°C (3.71%) and 60°C (5.54%). This can be supported by the findings in which 70ºC produced the highest pectin yield from grapefruit among the different temperatures (50, 60 and 70°C) investigated [13]. At the same time, it can be observed that the pectin yield is increasing with extraction time (sonication time) and reached the peak from 20 to 40 min. After the peak limit, the yield started to decrease until it became constant after 70 min. It is also noteworthy to report that the higher the temperature, the faster the peak yield was achieved before its decline. Previous studies also reported similar increasing extracted pectin yield by UAE with extraction time that ranged from 20 to 60 min [15, 21, 23, 26]. Enhanced extraction at high temperatures could be attributed to high solubility and thus improved the mass transfer. However, the negative impact of prolonged extraction was noted in this study. The reduction of pectin yield over extraction period may be the consequence of thermal degradation of the extracted pectin as high temperature can produce more cavitation bubbles [46]. This will increase the shear stress in order to break these bubbles. As a result, the pectin chain might degrade into smaller components leading to a decreased yield [13].

The temperature of 70°C was then selected as center point in CCD optimisation to produce different experimental conditions (as shown in Table 3) in order to find optimum condition for pectin extraction from studied fruit peel.

3.2. Optimisation of response surface methodology (RSM)

3.2.1. Experimental design

Optimisation of extraction process was performed using CCD in Design Expert software 7.0. Second-order quadratic model was employed to determine the effect of variables on the yield of pectin in this study. After the selection of the independent variables (temperature, time and LSR) and their respective ranges, using the experimental software with CCD, different combinations of extraction conditions were generated. Each run was performed thrice to acquire the average pectin yield. Table 3 represents the experimental design with the predicted and experimental result.

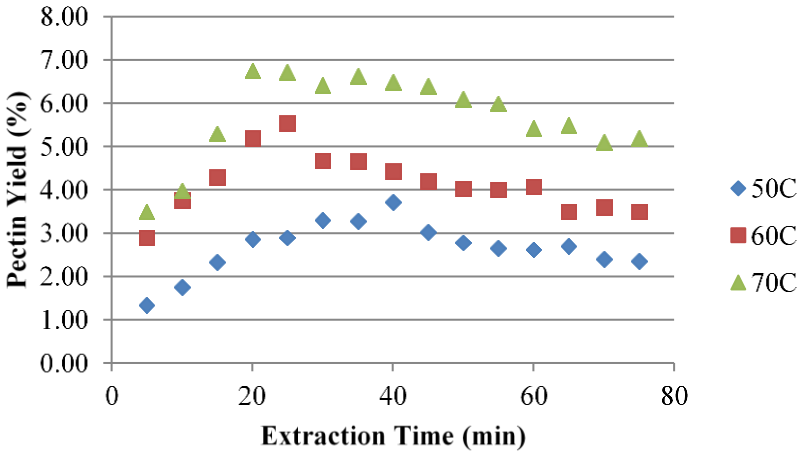

Fig. 1. Pectin yield against time.

Table 3. Central composite design (CCD) with experimental and predicted pectin yield.

Std	Run	Temp (°C)	Time (min)	LSR (mL/g)	Pectin Yield (%)	
					Experimental	Predicted
9	1	60	20	30	1.98	0.85
1	2	64	14	21	1.83	3.28
8	3	76	26	39	6.75	6.66
11	4	70	10	30	4.89	4.07
15	5	70	20	30	6.65	7.03
3	6	64	26	21	3.15	3.75
13	7	70	20	15	7.14	5.52
6	8	75	14	39	3.69	4.53
2	9	76	14	21	4.38	4.68
12	10	70	30	30	7.65	6.50
16	11	70	20	30	7.18	7.03
4	12	76	26	21	3.42	4.86
5	13	64	14	39	1.89	1.75
19	14	70	20	30	6.29	7.03
17	15	70	20	30	6.69	7.03
10	16	80	20	30	4.59	3.84
7	17	64	26	39	3.42	4.49
18	18	70	20	30	7.65	7.03
20	19	70	20	30	7.39	7.03
14	20	70	20	45	6.10	5.75

From Table 4, the experimental pectin yield ranged from 1.89 to 7.65%. Result showed the runs with extraction temperature of 70°C, extraction time of 20 min and LSR of 30:1 mL/g produced highest pectin yield (7.03%) whilst runs with temperature 64°C, time of 14 min and LSR of 39:1 ml/g produced the least pectin yield (3.28%). The effect of independent variables on the pectin yield was discussed in the following section.

3.2.2. Analysis of variance (ANOVA)

A multiple regression analysis with respect to the experimental results retrieved from CCD was performed. It was observed that a quadratic polynomial model produced a satisfactory fitting of the experimental data with regard to the total pectin yield. ANOVA was used to evaluate the adequacy of the model to exhibit the extraction of pectin from dragon fruit peel. The result in Table 4 showed that the quadratic model was able to explain most of the variation in the response due to high *F* value (*F* value = 5.0333) and low ρ-value (ρ= 0.0094) [16]. The final equation in terms of coded factors is presented in Eq. (12).

$$\begin{aligned}\text{Pectin Yield} = &-234.4883 + 6.60334X_1 + 0.64843X_2 - 0.17540X_3 \\ &- (2.03643 \times 10^{-3})X_1X_2 + (4.94096 \times 10^{-3})X_1X_3 \\ &+ 0.010508X_2X_3 - 0.046869{X_1}^2 - 0.017496{X_2}^2 \\ &- (6.21610 \times 10^{-3}){X_3}^2 \end{aligned} \quad (12)$$

The quality of the fitted model was checked by R^2, adjusted R^2 and adequate precision is shown in Table 4. High value of R^2 (0.8192) and adjusted R^2 (0.6564) indicated that the model could represent the actual relationship between independent variables and response (pectin yield). In this study, the value of R^2 implied that only 18.08% of the total variation could not be explained by the model [47]. Adequate precision measures the signal to noise ratio and it is desirable if the ratio is > 4 [48]. As shown in Table 4, the adequate precision of the developed model was 7.275, which showed the best fitness of the model.

Furthermore, the statistical significance of each variable was determined by *F*-value and ρ-value including the interaction between each variable. From Table 4, linear effect of extraction temperature (X_1), extraction time (X_2) and quadratic term of extraction temperature (X_1^2) were found as the significant model terms in this case (ρ < 0.05). The variable with larger *F*-value and smaller ρ-value indicated it had the most significant effect than others. Therefore, quadratic term of extraction temperature (X_1^2) had greater effect on the pectin yield followed by linear term of extraction temperature (X_1) and extraction time (X_2). The result demonstrated that the extraction of pectin was not significantly affected by linear term of LSR (X_3), quadratic effect of extraction time (X_2^2), quadratic effect of LSR (X_3^2) and interaction effect of these three independent variables (X_1X_2, X_1X_3 and X_2X_3) (ρ > 0.05).

3.3. Effect of variables on pectin yield

3.3.1. Effect of temperature on pectin yield

Extraction temperature significantly affected the extraction of pectin yield in this study. From Fig. 2, the pectin yield increased with increasing extraction temperature until 71.8°C but dropped with further increment of temperature. This phenomenon (pectin yield increased drastically at early extraction phase) happened because the solubility and diffusivity of solid from the plant material increased when temperature increased. Increased in temperature might cause disruption of the ester linkage and hydrogen bond which favoured the extraction process [15]. Nevertheless, solvent surface tension and viscosity might reduce with increased temperature. This might cause some disruptions to the characteristics of ultrasonic

cavitation and intensity of mass transfer enhancement [16]. Besides, thermal degradation of pectin could occur which led to further loss of pectin yield [13, 16].

Table 4. Analysis of variance (ANOVA) for the surface quadratic model.

Source	Sum of Squares	Degree of Freedom	Mean Square	*F*-value	*p*-value (*Prob* > *F*)
Model	65.3510	9.0000	7.2612	5.0333	0.0094
A-Temperature	10.5843	1.0000	10.5843	7.3367	0.0220
B-Time	7.1693	1.0000	7.1693	4.9696	0.0499
C-LSR	0.0645	1.0000	0.0645	0.0447	0.8368
AB	0.0411	1.0000	0.0411	0.0285	0.8693
AC	0.5449	1.0000	0.5449	0.3777	0.5525
BC	2.5704	1.0000	2.5704	1.7817	0.2115
A^2	39.3352	1.0000	39.3352	27.2661	0.0004
B^2	5.5557	1.0000	5.5557	3.8510	0.0781
C^2	3.5502	1.0000	3.5502	2.4609	0.1478
Residual	14.4264	10.0000	1.4426		
Lack of Fit	13.1008	5.0000	2.6202	9.8826	0.0126
Pure Error	1.3256	5.0000	0.2651		
Cor Total	79.7774	19.0000			
Std. dev.	1.20		R^2	0.8192	
Mean	5.14		**Adj-R^2**	0.6564	
***C.V.*%**	23.38		**Pred-R^2**	-0.3144	
Press	104.86		**Adequate Precision**	7.275	

3.3.2. Effect of extraction time on pectin yield

Pectin yield was significantly affected by extraction time. Fig. 2 showed that the pectin yield initially increased drastically but slowed down at later extraction phase. This result was in accordance to the previous study where the pectin yield was reported to slow down after longer period of extraction time [14]. Long extraction time was not preferred due to the possibility of pectin degradation, which led to reduced pectin yield. Previous finding stated that shorter extraction time could prevent pectin side chain from degradation [21]. Furthermore, the exposure of ultrasound for a long period was reported previously to cause pectin degradation [17].

It was reported that the yield increased initially due to the swelling and hydration of the material which caused by cavitation effect of ultrasound wave. Disruption of cell wall and penetration of solvent into the plant matrix were enhanced which improved the extraction process [16]. Nevertheless, decomposition and destruction of pectin could be happened if prolong the extraction time (prolong the exposure to ultrasound) which reduced the pectin yield [16]. These explanations justified that the pectin yield increased significantly at early phase of extraction but slowed down during the later phase.

3.3.3. Effect on liquid-solid ratio (LSR) on pectin yield

Although LSR did not significantly affect the pectin yield in this study, it was found that the pectin yield increased as the LSR increased up to 30 mL/g. The pectin yield was also noted to decrease when LSR was beyond 30:1 mL/g. The contact area between material and solvent increased when LSR was increased. This caused the

polysaccharide to fully dissolve out from the material. However, when the LSR was further increased (beyond 30:1 mL/g), the distribution of ultrasonic energy density in the solution decreased. As a result, polysaccharide yield decreased by hindering the dissolution of polysaccharide [34].

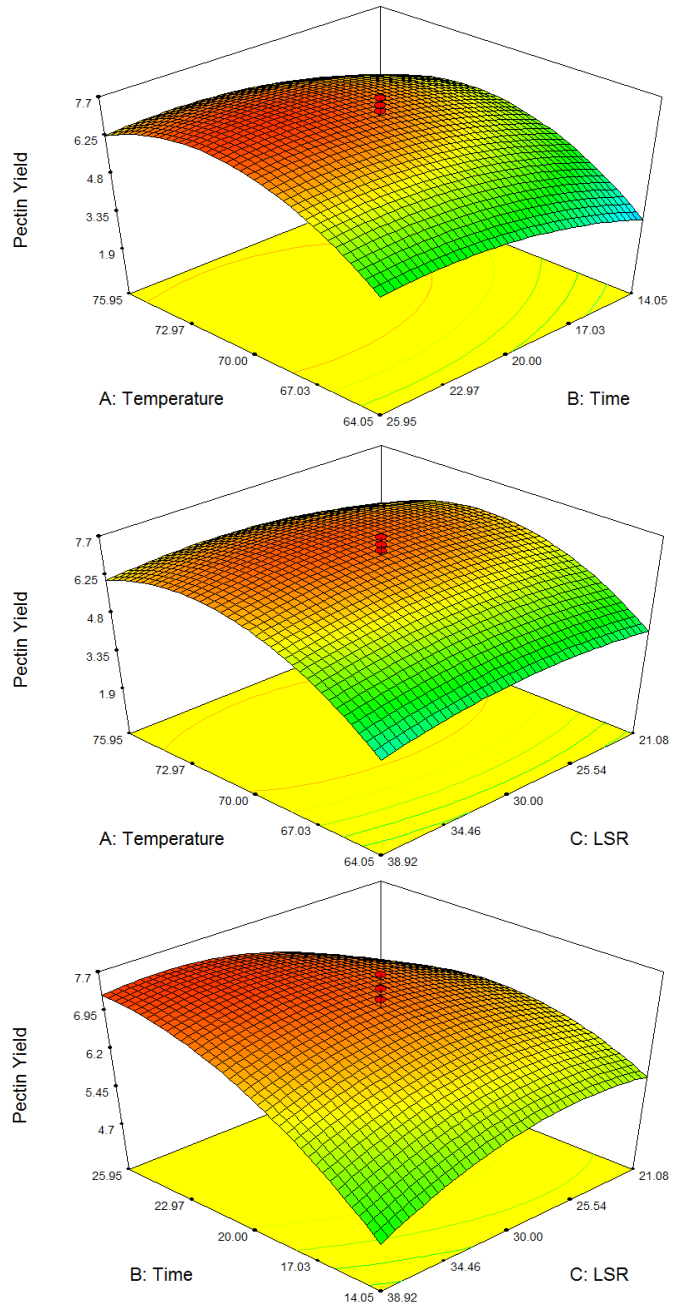

Fig. 2. Effect of extraction variables on pectin yield.

3.4. Determination and validation of optimum condition

Optimum condition for extraction to produce the highest pectin yield of 7.49% generated by Design Expert 7.0 was extraction temperature of 71.78°C, extraction time of 25.04 min and LSR of 35.57:1 ml/g with the desirability of 0.785. After performing the experiment thrice based on the optimum condition (as mentioned above) for validation, the actual experimental mean pectin yield acquired was 7.75% whereas the error percentage is 3.5%. The results showed that using RSM optimisation, the pectin yield had successfully improved by 0.26%.

3.5. Comparison of existing solid-liquid extraction (SLE) kinetic models

Table 5 shows the constants and coefficient of determination (R^2) of each of the kinetic models applied for the extraction temperature of 70°C. Peleg's Model was shown to be the most suitable model used to describe the extraction kinetics of pectin from dragon fruits as the coefficient of determination (R^2) value was the highest and closer to 1 among the models.

Table 5. Model's constant and coefficient of determination (R^2) of different models for solid-liquid extraction.

Model	Model's constants	R^2
First-order Kinetic model	k_1=0.1461	0.9537
Peleg's model	k_1=0.4305; k_2=0.1259	0.9642
Second-order rate equation	h=3.0745	0.9415
Logarithmic model	a=2.1186; b=3.8101	0.9256
Power Law equation	B=4.4573; n=0.1290	0.9177

4. Conclusions

In this study, UAE was a good approach to extract pectin from red dragon fruit peels. The result revealed that pectin yield was significantly affected by extraction temperature and time but not by LSR. Pectin yield increased with the extraction temperature and time. The application of RSM using CCD in this study had successfully developed an optimum condition with extraction temperature of 71.8°C, extraction time of 25 min and LSR of 35.6:1 mL/g with the maximum pectin yield of 7.49%. The condition was confirmed by validation experiments and ANOVA. The most suitable kinetic model that can represent the extraction of pectin from studied raw material was Peleg's model with the highest coefficient of determination (R^2) of 0.9642.

Although dragon fruit could be an ideal pectin source, there is still lacking of studies on the properties of pectin extracted from dragon fruit. Therefore, research on properties of extracted pectin from dragon fruit peels can be performed in the future and it is best if compared to other raw materials. In addition, optimisation process can be done not only based on the yield but also the properties of pectin for qualitative and quantitative purpose.

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Nomenclatures

C_t	Concentration of solute at any given time, mg solute/ g of solid material
C_o	Initial concentration of solute, mg solute/ g of solid material
C_α	Equilibrium concentration of solute, mg solute/ g of solid material
R^2	Coefficient of determination
X_1	Temperature
X_2	Extraction time
X_3	Liquid-solid ratio
W_p	Dried peel powder, g
W_t	Dried pectin mass, g

Greek Symbols

α	Code of parameter
ρ	Significance value

Abbreviations

ANOVA	Analysis of Variance
CCD	Central Composite Design
CV	Coefficient of Variance
DE	Degree of Esterification
DPPH	2,2-diphenyl-1-picrylhydrazyl
EV	Equal Volume
GA	Galaturonic acid
LSR	Liquid-Solid Ratio
MAE	Microwave Assisted Extraction
RSM	Response Surface Methodology
SLE	Solid-Liquid Extraction
UAE	Ultrasound-Assisted Extraction

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