

Optimisation of Extraction Conditions of Watermelon (*Citrullus Lanatus*) Rind using Response Surface Methodology

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Abstract. Watermelon white rind is considered the most underutilised fruit resource as it is usually discarded as waste due to its unappealing flavour. Utilising watermelon rind by generating new products could decrease the amount of biological waste, thus recognising the rind's potential economic value in various industries, mainly in the food and cosmeceutical industries. Therefore, this study aimed to determine the optimum conditions for extracting watermelon rind. The watermelon rind sample was dried using an oven before extracting the rind. Four factors were used: time, solvent concentration, solid-to-solvent ratio and drying time. These factors were set in Design-Expert version 13 to get the optimum conditions for the best extraction yield. The response surface methodology reveals that four conditions which are 30 mins extraction time, 90 % ethanol concentration, 40 % solvent-to-solid ratio, and 45 hrs sample drying time, showed maximum extract yield at 22 % when extracted using a sonication technique.

1 Introduction

Watermelon, *Citrullus lanatus* (Thunb.) refers to the family Cucurbitaceae, which includes several other fruits such as muskmelon, winter melon, squash, bottled ground, cucumber, and pumpkin [1]. The main parts of watermelon are flesh, seed and rind. [2] Reported that watermelon rind consists of 10 % (w/w) lignin, 13 % (w/w) pectin, 20% (w/w) cellulose and 23 % (w/w) hemicellulose. However, the percentage of the composition may vary depending on its genotype.

Watermelon rind is considered fruit waste in most regions due to its unappealing flavour. Watermelon rind can be used as high-fibre flour for baked foods [3]. L-citrulline from watermelon rind extract has been used as a dietary supplement in the pharmaceutical industry for the treatment of certain urea cycle disorders. [4]. In addition, watermelon rind

has been applied in wastewater treatment for the removal of heavy metals after being changed into a bio-sorbent material [2, 5].

Watermelon rind has been applied in various applications such as additives in food industries, supplements in the pharmaceutical industry and biosorbent material in wastewater treatment. The process of extracting compounds like antioxidants and pectin from watermelon rind was influenced by several factor conditions during the sonication process. Thus, there is a drive to search for appropriate sonication extraction conditions of watermelon rind by implementing Design Expert-12 experimental design which provides the optimum yield of extraction for industrial applications.

2 Methodology

2.1 Sample collection

The watermelon was purchased from the local Kelantan market. The fruit was in maturity index of 2 with 70 – 90 % ripped, medium size classification weighing 4 – 6 kg and free from physical defects. The morphology of the fruit was oval, with dark green skin with strips, yellowish ground spots, red flesh, and fewer seeds.

2.2 Sample preparation

Watermelon fruits were washed to remove any dirt from the skin. The fruit was cut diagonally on a clean preparation table to remove the red fleshy pulp. The hard-green skin also has been removed carefully. The rind was cut with 10 mm thickness into a cube and weighed. The watermelon rind was dried in a Memmert convection oven at 60 °C. Drying at 60 °C temperature was chosen considering that the watermelon rind contains about 90 % water content, and based on the reference procedure by Adegunwa et al. [3], Baeri et al. [6], Ho et al. [7], Lee & Choo [8], Naknaen et al., [9], Nurdalilah et al., [10], Petchsomrit et al., [11], Prakash Maran et al., [12], and Sanwiriya & Suleiman [13].

2.3 Optimization of extraction

Response Surface Methodology (RSM) using software Design Expert 12 (Stat-Ease, USA) was used to optimize the conditions for watermelon rind extraction. Four independent variables (solvent-to-solid ratio, ethanol concentration, sample drying time and extraction time) of each sample were taken to a three-factor level as shown in Table 1. A central composite design (CCD) consisting of 28 experimental runs was applied to evaluate the effect of variables, and the response was the yield of watermelon rind extract.

Table 1. Independent variables and factor levels for optimization of extraction.

Independent variables	Factor level		
	-1 (- α)	0	1 (+ α)
Ethanol concentration (%)	80	90	100
Solvent-to-solid ratio (v/w)	20:1	30:1	40:1
Extraction time (h)	30	45	60
Drying time (h)	42	45	48

2.4 Ultrasonic bath extraction

The extraction of watermelon rind was conducted using an ultrasound-assisted extraction method. Following the method conducted by Naknaen et al. [14] with modification, ethanol with watermelon rind sample was extracted in a flask placed in the R.S. Pro ultrasonic bath sonicator with an ultrasonic power of 100 W and frequency of 40 kHz. The aluminium foil was applied to cover the top of the flask to minimize the evaporation of the solvent to maintain the extraction ratio. The samples were centrifuged at 2,800 xg for 10 min using an Eppendorf centrifuge and filtered through 180 mm filter paper. The solvent was evaporated at 50 °C using a Heidolph rotary evaporator to obtain crude extract.

3 Result and discussion

Central Composite Design (CCD) was used to obtain the optimum extraction yield of watermelon rind extract subjected to oven and dehydrator drying. A total of 28 runs for each sample were conducted with four factors which are ethanol concentration (%), extraction time (min), solvent-to-solid ratio (%) and sample drying time (h), to three levels. All four responses of four independent variables are shown in Table 2.

The analysis of variance (ANOVA) for each variable generated a second-order polynomial model as per Equation 1.

$$Y_{\text{extraction yield}} = 379.13 + 0.0272A + 12.5B + 193.39C + 85.37D + 1.27AB + 3.52AC + 13.14AD + 1.27BC + 9.77BD + 8.27CD + 1.59A^2 + 29.22B^2 + 2.93C^2 + 6.07D^2 \quad (1)$$

Where Y is the predicted response; A is extraction time; B is ethanol concentration; C is solvent-to-solid ratio; and D is drying time.

As shown in Table 3, the model is significant, with an F-value of 10.74, indicating only a 0.01% likelihood that such a large F-value is due to random noise. P-values below 0.0500 signify that the model terms are important. In this case, the terms B, C, D, AD, and B² are significant. Conversely, P-values above 0.1000 suggest that the model terms are not significant. If several insignificant terms are present (excluding those necessary for maintaining hierarchy), reducing the model may enhance its performance. The Lack of Fit F-value of 0.53 shows that the Lack of Fit is not significant compared to pure error, with an 80.58% probability that such a high value could be attributed to noise.

The non-significant lack of fit is adequate for the model. The model's regression (R²) was 0.9204, with a standard deviation of 1.59, indicating a good model accuracy as nearly all data could be explained by the model as stated in Equation. The lack of fit F-value of 0.53 is not significant relative to the pure error.

Figure 1 shows that the yield of watermelon rind extracts efficiently increased as the extraction time and ethanol concentration increased. The highest yield (14.3 %) can be obtained at extraction conditions of 45 min extraction time, 80 % ethanol concentration, 30 % solvent-to-solid ratio, and 45 h sample drying time. The interactions between ethanol concentration, extraction time, sample drying time and solvent-to-solid ratio showed noticeable effects (p<0.05) on the watermelon rind extraction yield. The extraction yield increased by reducing the extraction time; increasing ethanol concentration and solvent-to-solid ratio. The result from this study exhibits a higher extraction yield which was 14.3 %, higher than Lee & Choo [15], as they only extracted 8.38 % watermelon rind pectin yield using ultrasound-assisted extraction. This phenomenon might be due to the different ultrasonic frequencies applied during extraction to rupture plant cell walls for solvent penetration.

Table 2. The response of the experimental run for oven drying sample

Run	Factor 1 Extraction time (min)	Factor 2 Ethanol concentration (%)	Factor 3 Solvent-to-solid ratio (%)	Factor 4 Drying time (h)	Response Yield (%)
1	45	90	30	48	8.3
2	45	90	30	45	11.3
3	45	90	30	45	9.8
4	60	100	40	48	10
5	45	100	30	45	12.8
6	30	90	30	45	10.5
7	60	100	20	48	7.5
8	30	100	20	48	5.5
9	30	80	20	42	12
10	60	100	20	42	10.5
11	30	80	40	48	14
12	45	90	30	45	12
13	45	90	20	45	6.5
14	45	90	30	42	9
15	60	80	20	42	10
16	60	100	40	42	17
17	60	80	20	48	11
18	30	80	20	48	6
19	45	80	30	45	14.3
20	45	90	30	45	7.5
21	60	80	40	48	15
22	60	80	40	42	18
23	45	90	40	45	16
24	30	80	40	42	19
25	30	100	20	42	11
26	30	100	40	42	20
27	30	100	40	48	10
28	60	90	30	45	8.3

Table 3. ANOVA of Quadratic model

Source	Sum of Squares	df	Mean Square	F-value	p-value
Model	379.13	14	27.08	10.74	< 0.0001
A-Extraction time	0.0272	1	0.0272	0.0108	0.9188
B-ethanol concentration	12.50	1	12.50	4.96	0.0443
C-Solvent-to-solid ratio	193.39	1	193.39	76.67	< 0.0001
D-Drying time	85.37	1	85.37	33.85	< 0.0001
AB	1.27	1	1.27	0.5018	0.4912
AC	3.52	1	3.52	1.39	0.2589
AD	13.14	1	13.14	5.21	0.0399
BC	1.27	1	1.27	0.5018	0.4912
BD	9.77	1	9.77	3.87	0.0708
CD	8.27	1	8.27	3.28	0.0934
A ²	1.59	1	1.59	0.6293	0.4419
B ²	29.22	1	29.22	11.58	0.0047
C ²	2.93	1	2.93	1.16	0.3008
D ²	6.07	1	6.07	2.41	0.1447
Residual	32.79	13	2.52		
Lack of Fit	20.90	10	2.09	0.5273	0.8058
Pure Error	11.89	3	3.96		
Cor Total	411.92	27			

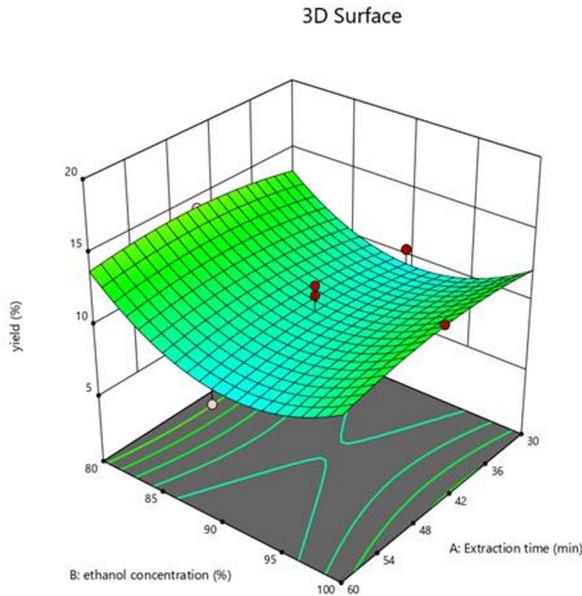


Fig. 1. Interaction between ethanol concentration (%) and extraction time (min) for optimising the extract yield

4 Conclusion

In conclusion, the study showed different extraction conditions can help in maximizing different watermelon rind sample conditions. The interactions between extraction time, ethanol concentration, solvent-to-solid ratio, and sample drying time showed noticeable

effects ($p < 0.05$) on the yield of watermelon rind extract. Lower extraction time, higher ethanol concentration and higher solvent-to-solid ratio showed an increment yield rate. The interactions could be applied to the extraction of other plants as well. Hence, the application of Response Surface Methodology has the potential to identify the most preferable conditions in extracting watermelon rind extract for large-scale industrial purposes.

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