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Original Research Paper

# Optimization of Pectin Extraction from Albedo of Watermelon (*Citrullus lanatus*) using Response Surface Methodology

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**Abstract:** This research delves into the utilization of albedo of watermelon rind as a potential source of pectin, a vital polysaccharide with wide-ranging applications in food and pharmaceutical industries due to its gelling and stabilizing properties. The study specifically focuses on the extraction of pectin from watermelon (albedo) using advanced methodologies like central composite design (CCD) and response surface methodology (RSM). These methods allow for the optimization of key processing parameters: temperature (at levels of 60, 80, and 100°C), pH (at levels of 1.5, 2.0, and 2.5), and extraction time (at levels of 60, 90, and 120 minutes). The study's findings highlight how these factors have a major impact on the amount of pectin that is extracted. The complex link between the input variables (temperature, pH, and extraction time) and the final pectin yield is thoroughly explained by means of a second-order polynomial model. Notably, the study identifies optimal conditions for pectin extraction, where the highest yield of 6.42% was achieved. These optimal conditions entail a temperature of 100°C, pH of 1.5, and extraction time of 60 minutes. Furthermore, the research highlights the individual impact of temperature, pH, and extraction time on pectin yield, demonstrating their significant roles in the extraction process. By providing insights into the optimization of pectin extraction from albedo of watermelon this study contributes to the broader understanding of utilizing agricultural by-products for value-added applications in various industries.

**Keywords:** Albedo of watermelon, extract parameters, pectin, RSM.

## Introduction

The middle wall of plant tissue and the principal cell wall lamella include the complex family of galacturonic acid polysaccharides known as pectin. Pectin is widely utilized in a variety of culinary, pharmaceutical, and cosmetic items as a gel forming and stabilizer. A hydrocolloid substance that is frequently utilized in the food and beverage sector is pectin. Pectin is able to form colloids and gels in solution so that it can be used as a stabilizer in the pharmaceutical and food industries. The Food and Drug Administration (FDA) classifies pectin and pectin modifications into the food group (Devianti et al., 2020).

The need for pectin in Indonesia tends to increase every year. To meet domestic pectin needs, Indonesia largely relies on imported pectin. It is projected that the need for pectin will reach 1,320.01 tons annually in 2020. Therefore, it is important to find sources of raw materials containing pectin (Devianti et al., 2020). Commercial pectin is made from sugar beet pulp, apple pulp, and citrus peels (lemon, lime, and orange). Currently, 85.5% of pectin in the global market is produced from orange peel, 14.0% from apple pulp, and the remaining 0.5% from sugar beet pulp, orange peel, lemon peel, lime peel, and orange peel with contributions of 56%, 30%, and 13% respectively (Ciriminna, et al., 2016).

During 2018–2023, the global pectin market is anticipated to expand by 7.3% (Picot-Allain et al., 2022). Citrus output is frequently impacted by pectin-related illnesses such canker, oil spot, and black spot, in addition to the growing demand for pectin worldwide (Sharif et al., 2018). Pectin has recently found usage in drug delivery, biopolymer, and biological applications (Nemiwal et al., 2021). Additionally, it has been employed as a green corrosion inhibitor (Núñez-Morales et al., 2022) and an adsorbent (Shahrin et al., 2021; Alipour et al., 2021). Thus, it becomes crucial and timely to look for other sources of pectin. Water, cellulose, hemicellulose, soluble sugars, essential oils (particularly D-limonene), and polyphenols (particularly flavonoids) make up the majority of the peel of grapefruits (Citrus maxima). Furthermore, grapefruit peel has a small amount of pectin, vitamin A, vitamin C, amylase, and peroxidase (130–170 mg/100 g fresh material) (Mamiru dan Gonfa, 2020).

Agricultural, post-harvest, processing, and distribution activities account for over 60% of all food waste, which results in major losses for farmers as well as issues with food security and the environment (FAO, 2019). With a global production of around 103 million tons in 2018, watermelon (Citrullus lanatus) is the second largest fruit crop in the world and a fruit of significant economic importance (FAOSTAT CROPS, 2019). About 30% of the fruit is watermelon rind (WR), which is frequently thrown away carelessly into the environment, leading to environmental issues (Méndez *et al.*, 2021). Raw material sources can be obtained from food waste that has no economic value so that it can increase its added value, including utilizing watermelon skin albedo waste. South Sulawesi is also a producer of horticultural crops, in addition to food crops, especially land for planting watermelon of 670 ha in 2021 with a production of 4685 kw (BPS, 2022).

Watermelon (*Citrullus lanatus*) production worldwide in 2020 was 101 million tons making it one of the most economically important fruits [1]. According to Ciriminna et al., (2016), watermelon is currently grown in 122 countries and on every continent. Approximately 30% of watermelon consists of rind and flesh, while 2% consists of seeds (Picot-Allain et al., 2022). One of the main solid wastes dumped into the environment is watermelon rind (Picot-Allain *et al.,* 2022). More than 90% of watermelon skin waste is disposed of as residue into the environment, making it an environmental challenge (Picot-Allain *et al.*, 2022). According to Sharif et al., (2018), approximately 36 million tons of watermelon skin were dumped into the environment in 2013. Despite numerous publications regarding the prospective uses of watermelon rind, its waste is not yet used for any industrial applications (Picot-Allain et al., 2022). Making use of this trash is crucial for both environmental and economic reasons. Carbs, proteins, lipids, minerals, vitamins, and more are all found in watermelon (Sharif et al., 2018). Since watermelon rind is primarily composed of carbohydrates, it can be utilized to extract useful substances like pectin (Mamiru dan Gonfa, 2023).

Samples containing various minerals and organic acids are the primary means of extracting pectin. Additionally, the raw materials and extraction conditions particularly pH, temperature, and extraction time have a significant impact on the characteristics of pectin (Belkkheire et al., 2019). Therefore, optimization of significant factors from the research results using Response Surface Methodology, this is intended to obtain better results and quality of the extracted pectin. Response surface methodology (RSM) is a powerful tool for designing, statistical modeling, and optimizing biological processes, and also for determining the relationship between one or more process variables.

## Material and Method

### Materials

Watermelon (*Citrullus lanatus*) fruit was obtained from a selection at the Pa'baeng-baeng traditional market in Makassar City, South Sulawesi as an experimental material in this study. The red portion of the fruit was removed during processing, leaving behind the white skin (albedo), which was then chopped into 0.5–2.5 cm pieces and gently stirred for 10 minutes in distilled water. The fruit peel was then dried in an oven set to 60 degrees Celsius for five days after the water was drained. Furthermore, the dried watermelon skin albedo sample was ground and sieved (Méndez *et al.*, 2021).

### Extraction of pectin

A total of 10 g of watermelon skin albedo powder was mixed with 400 ml of distilled water (1:40) and acidified with 0.5 N hydrochloric acid (HCl) to the pH, temperature and extraction time designed using the CCD - RSM method. The yield of pectin extract from albedo of watermelon was calculated as the dry pectin weight (g) per 100 g dry weight of watermelon rind albedo powder referring to (Pagarra *et al.*, 2019).

### Optimization of Pectin Extract Yield with CCD Method - Response Surface Methodology

Optimization experiment of watermelon rind albedo pectin extraction using *the Central Composite Design* method (*Response Surface Methodology*) to optimize the variables in the production of watermelon rind albedo pectin extract with pH variables (1.5 - 2.5), temperature (60 - 100) °C and extraction time (60 - 120) minutes (Pagarra *et al.*, 2019). The variables pH, Temperature and Extraction Time and the values of the variables are coded as -α, -1, 0, +1, + α (low, basal, high) can be seen in Table 1. The red portion of the fruit was removed during processing, leaving behind the white skin (albedo), which was then chopped into 0.5–2.5 cm pieces and gently stirred for 10 minutes in distilled water. The fruit peel was then dried in an oven set to 60 degrees Celsius for five days after the water was drained.

**Table 1.** Variable range and code value level

| Variable | Code Value | | | | |
|---|---|---|---|---|---|
| | **-∝** | **-1** | **0** | **+1** | **+∝** |
| pH | 1.16 | 1.5 | 2.0 | 2.5 | 2.84 |
| Temperature (ºC) | 46.36 | 60 | 80 | 100 | 113.64 |
| Extraction Time (min) | 39.55 | 60 | 90 | 120 | 140.45 |

## Result and Discussion

### *Optimization of Pectin Extract Results with Response Surface Methodology*

The determination of variables influencing pectin extraction, as revealed by the Two Level Factorial Design, emphasized the significance of pH, temperature, and extraction time (Palanikumar *et al.*, 2021; Bhattacharya, 2021; Paggara *et al.*, 2019). Subsequently, Central Composite Design (CCD) experiments were conducted to optimize these crucial variables in pectin production. Utilizing CCD, diverse combinations of pH and temperature were explored via the CD matrix, facilitating the establishment of a mathematical correlation model among significant variables. Moreover, optimal values for each factor and their interaction effects could be determined.

### *Experimental Design and Statistical Analysis*

The optimization of pectin extraction results was achieved through response surface methodology (RSM), focusing on the significant factors: pH (A), temperature (B), and extraction time (C). These factors were encoded as -α, -1, 0, +1, +α, representing low, basal, and high levels. RSM, along with CCD, standardized extraction parameters, with experimental pectin yield (%) values recorded (Table 2).

**Table 2.** Experimental values with different pH, temperature and extraction time for pectin yield with CCD by RSM

| Run | Factor A pH | Factor B Temperature (ºC) | Factor C Extraction Time (min) | Response Pectin Yield % |
|---|---|---|---|---|
| 1 | 1.50 | 100.00 | 120.00 | 6.16 |
| 2 | 1.50 | 100.00 | 60.00 | 6.42 |
| 3 | 2.00 | 80.00 | 90.00 | 5.41 |
| 4 | 2.00 | 80.00 | 90.00 | 5.27 |
| 5 | 2.50 | 100.00 | 120.00 | 4.84 |
| 6 | 2.00 | 80.00 | 90.00 | 5.26 |
| 7 | 2.00 | 80.00 | 140.45 | 3.75 |
| 8 | 2.00 | 80.00 | 90.00 | 5.01 |

| | | | | |
|---|---|---|---|---|
| 9 | 2.50 | 60.00 | 120.00 | 2.26 |
| 10 | 1.50 | 60.00 | 60.00 | 3.03 |
| 11 | 2.00 | 46.36 | 90.00 | 2.17 |
| 12 | 2.00 | 80.00 | 39.55 | 3.77 |
| 13 | 2.00 | 113.64 | 90.00 | 6.4 |
| 14 | 2.00 | 80.00 | 90.00 | 5.33 |
| 15 | 1.50 | 60.00 | 120.00 | 4.14 |
| 16 | 2.50 | 60.00 | 60.00 | 2.33 |
| 17 | 2.84 | 80.00 | 90.00 | 3.53 |
| 18 | 2.50 | 100.00 | 60.00 | 5.83 |
| 19 | 2.00 | 80.00 | 90.00 | 5.15 |
| 20 | 1.16 | 80.00 | 90.00 | 5.3 |

The results obtained, visualized in a 3D graph (Figure 1-3), demonstrate a rising trend in the watermelon rind's albedo pectin yield as temperature, pH, and extraction time increase. The impact of several parameters on the yield of pectin extraction was investigated using a three-dimensional response surface plot. These factors include solvent quantity, temperature and extraction time, as part of the RSM implemented on the Central Composite Design (CCD).

Extraction of pectin of albedo of watermelon rind involves a process using hot acid followed by precipitation with 96% ethanol. The yield of pectin was found to be greatly influenced by both the quality of the rind and the type of acid used. During the extraction process, 10 g of dry mass underwent extraction by adding hydrochloric acid with a pH range of 1.5 – 2.5, at different times and temperatures. Acids facilitate the hydrolysis of insoluble pectin constituents to soluble forms, thereby maximizing pectin recovery, especially under acidic conditions. On the other hand, increasing the pH level inhibits the release of pectin, possibly due to the aggregation of pectin. Furthermore, through experimental analysis, the probability value (P-value) for each parameter has been determined and provided in Table 3. This statistical analysis offers an insight into the importance of each parameter in influencing the pectin extraction results, helping in the determination of optimal extraction conditions.

**Table 3.** ANOVA for Response Surface Quadratic Model of Pectin Yield

| Source | Sum of Squares | DF | Mean Square | F Value | Prob>F |
|---|---|---|---|---|---|
| Mode | 34.83 | 91 | 3.87 | 77.09 | <0.0001* |
| A | 4.08 | 1 | 4.08 | 81.31 | <0.0001 |
| B | 25.34 | 1 | 25.34 | 504.79 | <0.0001 |
| C | 4.34 | 1 | 4.34 | 0.087 | 0.7746 |
| $A^2$ | 0.76 | 1 | 0.76 | 15.21 | 0.0030 |
| $B^2$ | 1.10 | 1 | 1.10 | 21.89 | 0.0009 |
| $C^2$ | 3.07 | 1 | 3.07 | 61.20 | <0.0001 |
| AB | 0.056 | 1 | 0.056 | 1.12 | 0.3153 |
| AC | 0.46 | 1 | 0.46 | 9.08 | 0.0130 |
| BC | 0.66 | 1 | 0.66 | 13.06 | 0.0047 |
| Residual | 0.50 | 10 | 0.050 | | |
| Lack of Fit | 0.40 | 5 | 0.081 | 4.06 | 0.0752 |
| Pure Error | 0.099 | 5 | 0.020 | | |
| CoreTotal | 35.34 | 19 | | | |
| Std Dev. | 0.22 | | $R^2$Square | 0.9858 | |
| Mean | 4.57 | | Adj.$R^2$ Square | 0.9730 | |

Values of "Prob > F" less than 0.0500 indicate model terms are significant (*)

Compared to other polynomial models, the response outcomes from this experiment that were examined using an ANOVA with a second-order polynomial model are more suitable. The regression equation that was produced is Equation 1. The following regression equation illustrates the connection between the variance test derived from RSM and the pectin extraction ratio:

Y (Pectin Yield) (%) = 05.23-0.55*A+1.36*B-0.018*C-0.23*A2-0.28*B2-0.46*C2+0.084*A*B- 0.24*A*C-0.29*B*C (1).

Here, Y represents the equation term for the coded factor. ANOVA confirmed model fit. A Prob > F value of less than 0.05 indicates the significance of the expression. When the p-value is less than 0.001, the term is also considered highly significant, indicating its large influence compared to other variables. This regression equation provides valuable insight into the relationship between the investigated factors and the resulting pectin yield, facilitating the identification of key parameters for optimizing the extraction process.

Since the coefficient of determination (R2) is high and around 1, the pectin yield determined by the second-order regression model is satisfied. The factor is considered important when the probability value is less than 0.05. This model has a probability of less than 0.0001, making it significant. The model is consistent with the experimental data, according to the regression coefficient, R2 value of 0.9858. By building a three-dimensional surface plot using the provided mathematical model equation, the ideal level for each variable element was identified. This indicates that the model explains 98.58% of the total variation. Table 3 displays the analysis of variance (ANOVA) results. For the extraction findings of watermelon rind albedo pectin, an ANOVA showed that the model agrees with the experimental data. The ideal extraction parameters were determined to be pH 1.5, temperature 100°C, and extraction time 60 minutes after each variable's relevance and significance were examined and evaluated. This produced an anticipated pectin yield of 6.42%. This, it can be concluded that pH, temperature, and extraction time are important factors in optimizing the extraction process. Its statistical significance underlines its crucial role in maximizing the yield of pectin extracted from watermelon rind albedo.

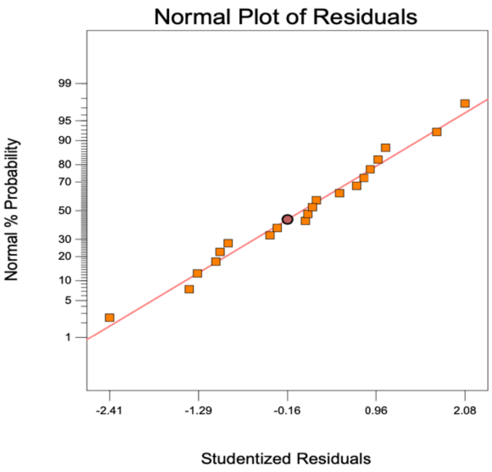

Fig. 1A

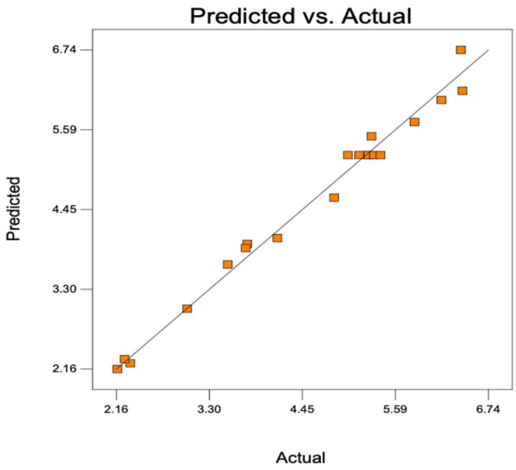

Fig. 1B

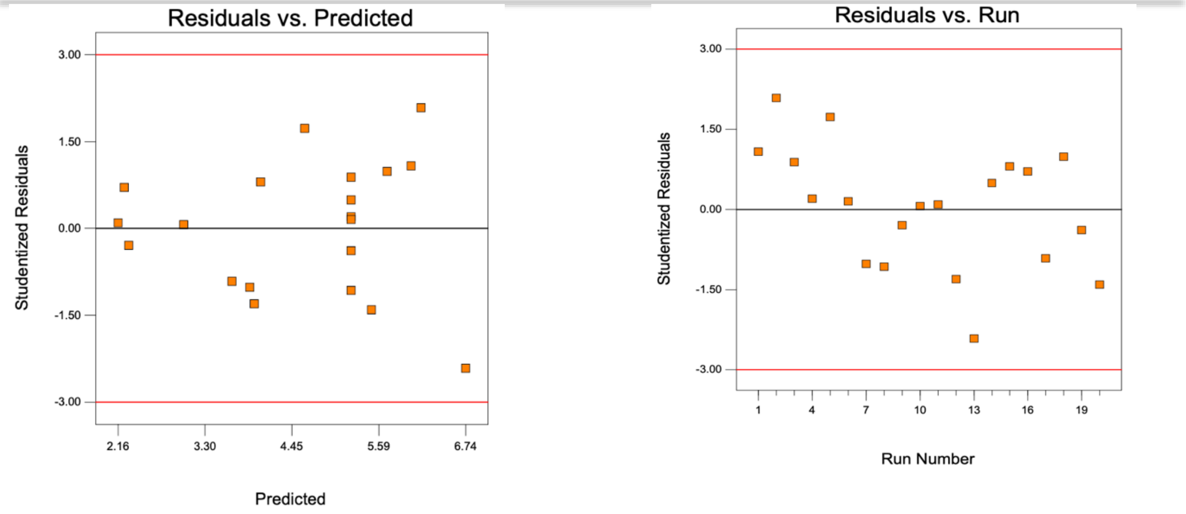

Fig. 1C

Fig. 1D

**Figure 1** A-D. Diagnostic plots: predicted against actual, normal percentage probability, and internally studentized residuals were used to assess the model's fitness.

Figure 1 (A-D) shows the assessment of the Adequacy of the model in addition to the coefficients by determining various indicative plots, including the percentage of normal probability, actual predictions, and internally learned residuals, which were made. Figure 1A shows the Percentage of Probability. Figure 1B shows the predicted value of the model is quite close to the investigated value, located close to the straight line, and shows a sufficient level of agreement with the actual data.

Figure 1C and 1D which show that these Residuals are consistent with the normal distribution because they are close to the straight line and do not show deviations, which is more predicted by the plot most of the data points are within the accepted statistical range. The 3-dimensional response surface images for the results are given in Figure 2 – Figure 4, which show the effects of A (pH), B (temperature) and C (extraction time) determined.

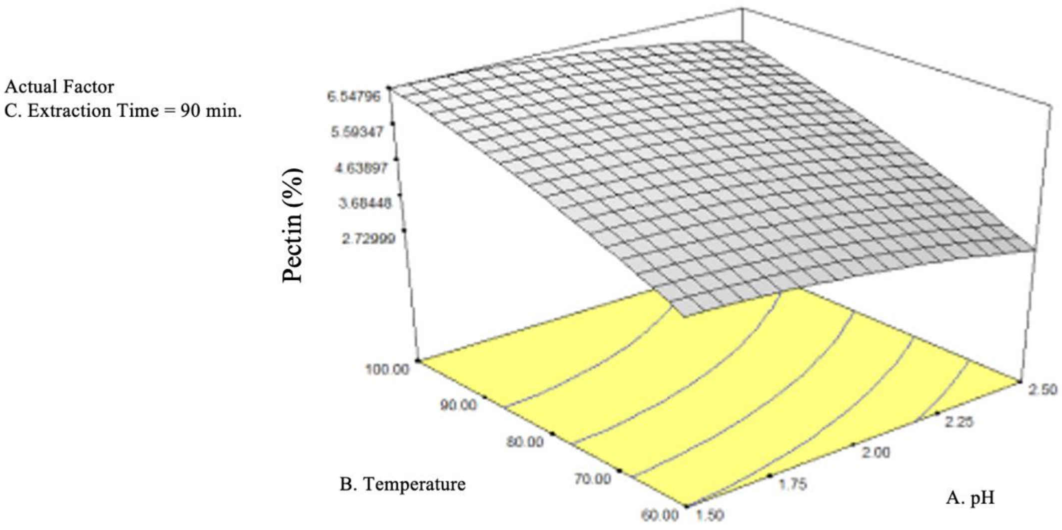

**Figure 2.** 3-Dimensional RSM for pectin yield as a function of pH and Temperature (Extraction time 90 minutes)

Figure 2 shows the results of watermelon rind albedo pectin extraction using the Response Surface Methodology with influential variables, namely pH and temperature, with the extraction time constant value at the midpoint (90 minutes). The effect of temperature on the response of watermelon rind albedo pectin produced in this study was quite significant, where the interaction between the temperature and pH variable factors showed a significant effect. Pectin from cotton banana peel was obtained by extraction using HCl solvent with pH 2.5 at a temperature of 100°C for 120 minutes. The yield obtained was 8.67% (Tuslinah *et al.*, 2023).

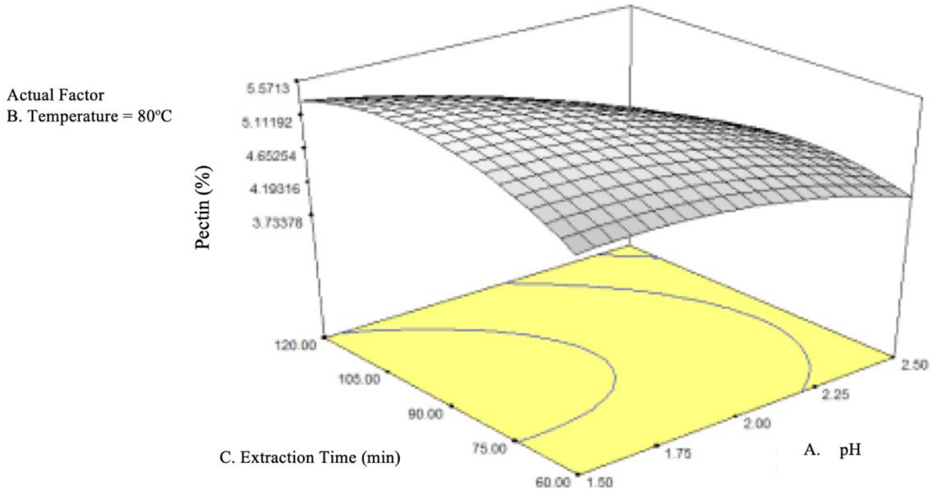

**Figure 3.** 3-Dimensional RSM for pectin yield as a function of pH and Extraction time (Temperature 80ºC)

The results showed that the interaction between pH and extraction time had a significant effect on the yield of grapefruit peel pectin extract. Figure 4 shows that the yield of pectin extract can increase and decrease within a certain range, depending on the pH level and extraction time used, while maintaining the extraction temperature of 80°C. The optimum yield of grapefruit peel pectin was achieved at pH 1.5, in accordance with the pH used in pectin extraction, which produced a significant yield of lemon peel (Yousuf *et al.*, 2018; Patil *et al.*, 2022) of 20.92% (pH range: 1, 1.5 and 2), and an extraction time of 30 minutes and a pH of citric acid solution of 1.5. Also reported that the highest pectin yield from sweet potato peel was 2.59%, achieved at a solution pH of 1.5, an extraction temperature of 90°C and an extraction time of 60 minutes (Yousuf *et al.*, 2019).

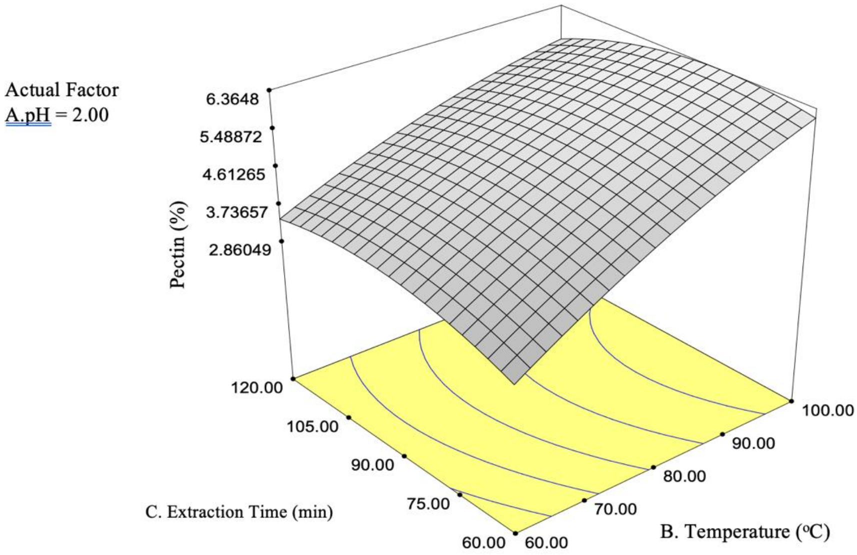

**Figure 3.** 3-Dimensional RSM for pectin yield as a function of Temperature and Extraction time (pH 2.00)

The results of the study showed that the extraction time and temperature on the production of watermelon rind albedo pectin with a constant pH at the midpoint (pH 2.0), are shown in Figure 4. The production of watermelon rind albedo pectin is depicted as a function of extraction time and temperature, highlighting a significant interaction effect between these variables. Pectin production tends to increase with longer extraction times and higher temperatures. Other studies have shown that pectin production from durian rind by extraction with temperature variations of 60oC, 80oC and 100 oC, and variations in extraction time, namely 30, 60 and 90 minutes. Durian rind was extracted using hydrochloric acid (HCL) and technical alcohol precipitant solution. In this study, the optimum conditions obtained were at a temperature of 80oC and a time of 60 minutes. The pectin obtained yielded 25.63% (Amanati, 2021).

## Conclusion

Pectin extraction from watermelon rind albedo was optimized using Response Surface Methodology (RSM), focusing on extraction temperature, pH, and time. The optimal conditions were found to be at 100°C, pH 1.5, and extraction time of 60 minutes, resulting in a maximum yield of 6.42%. The developed model showed high reliability, with predicted and actual results in good agreement. Significant interactions between extraction variables were observed, emphasizing their influence on pectin extraction. Overall, this study confirms the importance of these factors in maximizing pectin yield from watermelon rind albedo while maintaining quality within acceptable standards.

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