OPTIMIZATION OF THE AQUEOUS EXTRACT PROCESS FROM MANGOSTEEN PEEL FOR DYEING SILK

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ABSTRACT

This research studied the juice extraction process from mangosteen peel with water solvent and the parameters influencing on extraction process such as extraction temperature, extraction time and extraction ratio, and evaluating the juice of extract by determining extraction efficiency, color saturation of fabric and color fastness of silk fabric. The research results showed that the juice extracted from mangosteen peel can be used to dye silk fabric with high color strength and fastness. And the results of research will bring the efficiency of environmental, economy and society; and will meet urgent demands today.

Keywords: silk, natural color, mangosteen peel, extracting and testing a natural plant dye.

1. INTRODUCTION

Garcinia mangostana Linn is one of the local fruits of Viet Nam and the South Eastern area. Magosteen Rind has been used as a natural dye in textile applications so the component of the aqueous extraction of mangosteen contains a variety of compounds chromophore such as gartanin, BR-xanthone A, 3-isomangostin, α-mangostin, garcinon D… and other compounds. Moreover, the advantage of mangosteen peel is part of waste materials and mangosteen peel occupy 68 ~ 70% per mangosteen fruit [1, 2].

2. MATERIALS AND METHODOLOGY

2.1. Materials
Optimization of the aqueous extract process from mangosteen peel for dyeing silk

Silk fabric used was purchased from Nha Xa village, Moc Nam, Duy Tien, Ha Nam province, Vietnam; Silk fabric had fiber density of 40 g/m², brightness L = 88.65. Mangosteen peel was collected from Lai Thieu, Binh Duong from March to August every year. Mangosteen peel was ground into small particles from 0.5 to 1 mm, moisture content 8.5 %. The extracts of Mangosteen peel had been preserved in Kalisortbate 1 % at room temperature for 2 weeks.

2.2. Methodology

Effect of the aqueous extract process from mangosteen peel on silk fabric: The aqueous of mangosteen peel was extracted by water extraction and distillation. Investigated extract temperature was from 50 to 70 °C in 1 hour with ratio of mangosteen pulp/water of 1/5; the extract time were observed from 120 to 180 min at 80°C extraction for ratio of mangosteen pulp/water is 1/5; and the ratio of mangosteen pulp/water was extracted from 1/4 to 1/6 in 1 hour 30 minutes at 80 °C [1, 2, 3].

Dyeing silk fabric with aqueous extract from mangosteen peel: aqueous extract of mangosteen was mixed with water at the ratio of volume; heated up with the speed of 2÷3 °C/minute to 70 °C. The color intensities of dyed silk fabrics were determined on Minolta CR 300 colorimeter, Japan 73180167. The color fastness of dyed silk fabrics were tested by the standard of washing (ISO 105-C01) for the highest fastness levels 5 and the lowest fastness levels 1 [4]; and the properties of aqueous extracts and dyed silk fabrics were evaluated by spectroscopic analysis methods such as FT-IT, LC-MS and scanning electron microscope SEM.

Factorial experimental design of the extraction process from mangosteen rind: The model selected for this study is orthogonal center of complex pattern. Three variables were selected to study as extraction temperature (°C), extraction time (minutes), extraction ratio (mangosteen rind/water). Y1 is extraction efficiency and Y2 is color saturation of fabric, Xi and Xj represents the variables or parameters: bj, bji and bjj characterize the impact of variables Y1 and Y2, which showed by magnitude and sign of coefficients. Each value of objective function will be used to build a mathematical model in which a correlation between objective functions influencing factors was studied, given the form of polynomial equation [5]:

\[ Y = b_0 + \sum_{i=1}^{2} b_i X_i + \sum_{i=1}^{3} \sum_{j=1}^{2} b_{ij} X_i X_j + \sum_{i=1}^{2} b_{jj} X_j^2 \] (1)

3. RESULTS AND DISCUSSION

3.1. Building experiment models and regression equation for extraction conditions

Building experimental matrix: Between the variable coding (Xi) and the real variable value (Zi,), choosing to study at different levels are interrelated via the following formula:

\[ X_i = \frac{Z_i - Z_{0i}}{\Delta Z_i} \] (2)

in which: 

\[ Z_{0i}^{P}: \text{Research value at 0 (rootlevel)} \]

\[ \Delta Z_i: \text{The range of variables studied, } \Delta Z_i = \frac{Z_{\text{max}} - Z_{\text{min}}}{2} \] (3)
$Z_{\text{max}}$: The value study at high level (+1)

$Z_{\text{min}}$: The value study at low (–1)

With orthogonal plan, $\alpha$ value arm of the matrix is determined by the formula (4) with investigate levels of three selection variables (Table 1) and encoding matrix of three selection variables (Table 2)

$$\alpha = \sqrt[3]{\sqrt[3]{2^3 - 2^3} - 2^{3-1}} = \sqrt[3]{1.353} \quad (4)$$

| Investigate Variables (Real Variables) | Encoding Variables | Unit                      | Investigate Levels |
|---------------------------------------|--------------------|---------------------------|-------------------|
| Temperature ($Z_1$)                   | $X_1$              | °C                        | -1  0             |
| Time ($Z_2$)                          | $X_2$              | min                       | 120 150 180       |
| Ratio ($Z_3$)                         | $X_3$              | % the volume of water in the extraction mixture | 75 80 85 |

| Experiments | Encoding Variables | Objective Function |
|-------------|--------------------|--------------------|
| $n=2^3=8$   |                    | Extraction Efficiency ($Y_1$) | Color saturation of fabric ($Y_2$) |
| 1           | -1.0               | -1.0               | -1.0               | 8.42249 | 22.6309 |
| 2           | 1.0                | -1.0               | -1.0               | 13.8845 | 26.0337 |
| 3           | -1.0               | -1.0               | -1.0               | 765957  | 20.1894 |
| 4           | -1.0               | -1.0               | -1.0               | 11.9843 | 25.8653 |
| 5           | -1.0               | -1.0               | -1.0               | 9.23277 | 24.7244 |
| 6           | -1.0               | -1.0               | -1.0               | 13.8958 | 26.2337 |
| 7           | -1.0               | -1.0               | -1.0               | 9.85443 | 24.0709 |
| 8           | -1.0               | -1.0               | -1.0               | 11.2249 | 25.5362 |

| $n=2^2=4$   |                    | Extraction Efficiency ($Y_1$) | Color saturation of fabric ($Y_2$) |
| 9           | -1.353             | 0                 | 0                   | 7.23277 | 20.2164 |
| 10          | 1.353              | 0                 | 0                   | 11.5572 | 25.7254 |
| 11          | 0                  | -1.353            | 0                   | 10.5714 | 23.8362 |
| 12          | 0                  | 1.353             | 0                   | 11.1398 | 24.5147 |
| 13          | 0                  | 0                 | -1.353             | 19.2817 | 27.9945 |
| 14          | 0                  | 0                 | 1.353              | 20.915  | 28.8362 |

| $n=3$       |                    | Extraction Efficiency ($Y_1$) | Color saturation of fabric ($Y_2$) |
| 15          | 0                  | 0                 | 0                   | 23.839  | 30.2588 |
| 16          | 0                  | 0                 | 0                   | 23.7057 | 30.358  |
| 17          | 0                  | 0                 | 0                   | 23.399  | 30.3856 |
Table 3. Results of extraction efficiency and color saturation fabric by orthogonal matrix level 2.

| Exp. | X₁  | X₂  | X₃  | Temp. (°C) | Time (min) | Ratio (%) | Extraction Efficiency Y₁ | Color Saturation of Fabric Y₂ |
|------|-----|-----|-----|-----------|------------|-----------|--------------------------|-----------------------------|
| 1    | -1  | -1  | -1  | 50        | 120        | 75        | 8.42249                  | 24.6309                     |
| 2    | 1   | -1  | -1  | 70        | 120        | 75        | 13.8845                  | 24.9337                     |
| 3    | -1  | 1   | -1  | 50        | 180        | 75        | 7.65957                  | 25.6894                     |
| 4    | 1   | 1   | -1  | 70        | 180        | 75        | 11.9843                  | 25.8653                     |
| 5    | -1  | -1  | 1   | 50        | 120        | 85        | 9.23277                  | 24.0244                     |
| 6    | 1   | -1  | 1   | 70        | 120        | 85        | 13.8958                  | 24.6337                     |
| 7    | -1  | 1   | 1   | 50        | 180        | 85        | 9.85443                  | 24.4709                     |
| 8    | 1   | 1   | 1   | 70        | 180        | 85        | 11.2249                  | 25.8362                     |
| 9    | -1.353 | 0   | 0   | 46.47     | 150        | 80        | 7.23277                  | 24.8164                     |
| 10   | 1.353 | 0   | 0   | 73.5      | 150        | 80        | 11.5572                  | 25.8254                     |
| 11   | 0   | -1.353 | 0   | 60        | 117.06     | 80        | 10.5714                  | 25.8362                     |
| 12   | 0   | 1.353 | 0   | 60        | 182.94     | 80        | 11.1398                  | 25.8147                     |
| 13   | 0   | 0   | -1.353 | 60        | 150        | 73.235    | 19.2817                  | 26.9945                     |
| 14   | 0   | 0   | 1.353 | 60        | 150        | 86.765    | 20.915                   | 25.8362                     |
| 15   | 0   | 0   | 0   | 60        | 150        | 80        | 23.839                   | 27.2588                     |
| 16   | 0   | 0   | 0   | 60        | 150        | 80        | 23.7057                  | 27.358                      |
| 17   | 0   | 0   | 0   | 60        | 150        | 80        | 23.399                   | 27.3856                     |

The maximum values of two objective functions be used to build mathematical models (1). Extraction efficiency and color saturation of fabric obtain from experiments (Table 3). From the results of color saturation of fabric and software Statgraphics Centurion XV.II, carried out statistical analysis correlation and regression to determine the regression of the process, checked conformity of the mathematical model to find the optimal conditions for extraction extracts from mangosteen peel. Then carried out analyze and appreciate the influence of the investigate variables to extraction efficiency of the extraction process and color saturation of fabric, and determine the optimal variables of the process.

**Analysis of variance and regression:** The Pareto Fig. 1 showed that the effect of investigate variables to extraction efficiency of the extraction process and color saturation of fabric in which the letters A, B, C represent encoding variables: X₁, X₂, X₃. The impact level of investigate factors of the extraction process was assessed by statistical analysis of experimental results. The column value showed the level of impact of factors quadratic and interaction of two variables simultaneously. The gray lines showed interaction effects on color saturation of fabric are positive effects (+) which increases extraction efficiency and color saturation of fabric. The blue lines are negative effects (-) which reduce extraction efficiency and color saturation of fabric. The value ratio of F is high and index P is low, the regression coefficients are more meaningful and more significant impact.
Thus, Fig. 1 showed that the effect of factors type 1 (A, B, C) and interactive effects level 2 AA, BB, CC are substantial and are retained so F is high and P-unreliability value is lower 0.05 (ie. Reliability ≥ 95 %). The residual effects which no meaning were removed because P is higher 0.05.

Table 4. The estimated value of regression coefficients.

| Factors | Regression coefficients | The value of regression coefficients |
|---------|-------------------------|-------------------------------------|
|         |                         | Extraction Efficiency (Y₁) | Color Saturation of Fabric (Y₂) |
| Const   | b₀                     | -526.414                  | -232.439                  |
| A: temp. | b₁                    | 8.7853                    | 4.87044                   |
| B: time | b₂                    | 1.91145                   | 0.664662                  |
| C: ratio | b₃                    | 3.3449                    | 1.4236                    |
| AA      | b₁₁                   | -0.0634884                | -0.0308128                |
| AB      | b₁₂                   | -0.00184578               | 0.000928792               |
| AC      | b₁₃                   | -0.00876112               | -0.0142486               |
| BB      | b₂₂                   | -0.00616786               | -0.00269266               |
| BC      | b₂₃                   | 0.00047768                | 0.000979536               |
| CC      | b₃₃                   | -0.0175431                | -0.00375685               |

Data processing by Statgraphics software obtained the regression with real variables (Z) và encoding variables (x) in Table 4. The objective function of Extraction Efficiency:

\[
Y₁ = -526.414 + 8.7853Z₁ + 1.91145Z₂ + 3.3449Z₃ - 0.0634884Z₁^2 - 0.00184578Z₁Z₂ - 0.00876112Z₁Z₃ - 0.0047768Z₂Z₃ - 0.0175431Z₃^2
\]

The objective function of Color Saturation of Fabric:

\[
Y₂ = -232.439 + 4.87044Z₁ + 0.664662Z₂ + 1.4236Z₃ - 0.0308128Z₁^2 + 0.000928792Z₁Z₂ - 0.0142486Z₁Z₃ - 0.00616786Z₂Z₃ - 0.000979536Z₂Z₃ - 0.00375685Z₃^2
\]

Check the suitability of model by the standard of Fisher (f₀ₙ) và Student (t₀ₙ) with reappear variance S²₀ₙ is calculated based on the central experiments n₀ = 3 and significance level α = 0.05, reappear freedom degrees f₀ₙ=3-1=2. The correlation coefficient value (R²=95.701)
indicating that 95.701 % variations of extraction efficiency are due to impact of independent variables and 4.299 % are due to external factors which unexplained by the model. Likewise, the correlation coefficient value ($R^2=92.3481$) indicating that 92.3481 % variations of Color Saturation of Fabric are due to impact of independent variables and 7.6519 % are due to external factors which unexplained by the model. This showed that the significance of of the two objective functions is rather high.

3.2. Analyze level affecting of factors to extraction efficiency and color saturation of fabric

![Figure 2. The influence of the investigate factors to extraction efficiency (a) and color saturation of fabric (b).](image)

**Effect of extraction temperature to extraction efficiency and color saturation of fabric:** The extraction temperature factor contributeing to promote the process of dissolving of the substances in mangosteen peel and impact mainly to extraction efficiency and color saturation of fabric (Fig. 2). Color saturation of fabric increases with increasing temperature and reached the highest value at 60 °C. Temperatures bring positive effects to increase in temperature range 60-70 °C. When temperature increase 70 °C then color saturation and intensity decrease reflected some regression level 2 ($b_{11}$).

**Effect of extraction time to extraction efficiency and color saturation of fabric:** Extraction time has a positive and significant impact to extraction efficiency and color saturation of fabric but lower. Time factor affecting level 2 to color saturation of fabric reflected regression coefficient $b_2$ in equation (5 and 7). Color saturation of fabric is obtained maximum value at 150 minutes; the study showed it is about time enough to completely dissolve the substances in mangosteen peel in extraction process. As the temperature factor is higher when the time 150 minutes, the color intensity reduction reflected by the regression coefficient level 2 ($b_{22}$).

**Effect of extraction ratio of extraction efficiency and color saturation of fabric:** The results of analysis showed that the percentage discount services can also influence the intensity level 2 to color fabric, but the effect of extract ratio decreased color intensity fabric and bring negative effects resulting from the regression equation (5 and 7) to the value of coefficient $b_3$. In this study, with extraction ratio 1/4-1/6, extraction efficiency and color saturation of fabric increased and obtained the maximum value in center survey then decrease; this is due to extraction ratio increasing, excess water will dilute some chromophore compounds in mangosteen peel powder.

**Result optimization:** From the regression equation obtained, optimization conditions mangosteen extract by software Statgraphics. Conditions of optimal extraction have received: color saturation of fabric 30.08; extraction temperature at 61.4 °C; extraction time 149.2 minutes and
84.6342 % mixed water respectively with extraction ratio 1/5. Carried out dye silk fabric with the optimal extraction process, check the color fastness indicators are achieved level 4-5/5.

### 3.3. Reviewed optimization results

![FT-IR Spectroscopy and Chromatography Coupled Mass Spectrometry](image)

**Figure 3.** The results of FT-IR infrared spectroscopy and chromatography coupled mass spectrometry MS of extracts before and after dyeing.

![SEM Results](image)

**Figure 4.** SEM results of silk fabric before and after dyeing with extracts from mangosteen peels.

The FT-IR spectral results in Figure 3 (a) and 3 (b) found that absorption spectrum appears chemotherapy ranged OH, C=C; C=O, a perfect fit with linking group of mangosteen ingredients in prior studies [1,2,3]. On the other hand, after dyeing with extracts, there are several slight differences from previous dye extracts, but at the peak change specific to chromophore (2102 cm⁻¹; 1284; 1085; 1044 cm⁻¹) so these groups have mounted color on canvas. At the same time, the results capture MS spectrum of optimized extracts in Figure 3 (c) and 3 (d) have also noticed the appearance of a lot of components in the extract chromophore as α-mangostin; β-mangostin; γ-mangostin (M = 409.3 g/mol), Acid gallic (M = 169.3 g/mol), Gartanin (M = 396.45 g/mol), 3-isomangostin (M = 409.3 g/mol), cyanidin (M = 288.4 g/mol), Xanthone (M = 196.9 g/mol).

And after dyeing service spectrum we can see loss of some compounds as α-mangostin, β-mangostin, γ-mangostin, acid gallic, 3-isomangostin, this can be confirmed that compounds of chromophore have linked on silk fabrics.

**On dyed silk fabric:** The SEM results in Figure 4(a) and 4(b) of silk fabrics before and after dyeing with extracts from mangosteen peel at the same resolution showed appearance of some part tiny particles bind and fill the small slot on fiber surface; while there is a thin film on fiber after dye surface, fabric before dye is none. This proves that the chromophore element linked on silk fabrics makes surface structure of silk fabrics changes.
4. CONCLUSION

The results of optimum extraction conditions were obtained temperature at 61.4 °C, time 149.2 minutes and percent of extract ratio is 84.6342 % (1/5). The results showed the juice extracted from mangosteen peel can be use to dye silk fabric with high color strength and fastness. The results allow hope a dyeing natural colors technology were produced in industrial model, can utilizing waste material, solve environmental problems towards green technology and sustainable development.

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