Optimization of Green Extraction Process of *Cinnamomum camphora* Fruit Dye and its Performance by Response Surface Methodology

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Natural dyes are widely available and mainly extracted from natural plants with green, safe, and sustainable characteristics. This study used *Cinnamomum camphora* fruit peel as the raw material under the premise of minimizing chemical reagents and employed the microwave-extraction method to extract the dyes from *C. camphora* fruit peel. The dye extraction conditions were optimized using response surface software *via the* Box-Behnken model to formulate a response surface test protocol. The results showed that optimizing the experimental conditions by the response surface method was reliable. The optimal extraction condition was found to be material-liquid ratio 1:20 (g: mL), microwave time 90 s, and microwave power 420 W. It is of practical application value to improve the extraction quantity of *C. camphora* fruit dye. At the same time, the infrared spectrum and HPLC-MS analysis of *Cinnamomum camphora* fruit dye were analyzed, and the stability of the dye was tested.

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Keywords: Cinnamomum camphora; Fruit peel; Response surface methodology; Natural dyes; Microwave-assisted extraction

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INTRODUCTION

In recent years, there has been an international wave of awareness about the environment, ecology, and pollution management, and there is a growing interest in using more environmentally friendly products (Fathordoobady *et al.* 2019; Fu and Zhu 2021). Natural dyes derived from plants have significant advantages over synthetic dyes (Zhang *et al.* 2021). Because of their non-toxic and biodegradable nature, the natural dye waste streams are increasingly valued for their ease of disposal compared to synthetic dye waste streams (Goodman *et al.* 2018; Yan *et al.* 2020; Kumar *et al.* 2021; Hu *et al.* 2022). The use of natural dyes for dyeing has a long history, but there needs to be more scientific research in many aspects (Liu *et al.* 2020, 2021a; Yan and Chang 2020; Qingqing *et al.* 2021, 2022). Natural dyes come from a wide range of raw materials, are biodegradable, green, and pollution-free. Their eco-friendly nature also depends upon their reasonable development, conducive to the protection of natural resources and ecological environment and meeting the requirements of sustainable development. At present, synthetic chemical dyes are mostly used for wood dyeing. Excessive use of such synthetic dyes will potentially threaten human health and the natural environment. The wood is dyed to enrich the color of the wood, make the color of the wood more uniform, to meet the requirements of the decorative wood. Studies have shown that natural dyes extracted from plants can be used to color and protect wood from threats. Some natural dyes can impart antidegradative and insect-proof properties to the material being dyed, indicating that the use of natural dyes to achieve the protection of materials is also an important aspect (Yeniocak *et al.* 2018; Feng *et al.* 2019; Xie *et al.* 2020; Li *et al.* 2021; Liu *et al.* 2021b; Yu *et al.* 2021).

There are abundant extraction methods for natural dyes, such as solvent extraction, microwave extraction, ultrasonic extraction, supercritical extraction, *etc*. The solvent extraction method often uses an organic solvent as the extraction medium, which is relatively simple. However, the extraction temperature typically is relatively high, as is the amount of solvent consumed, and the time required is long. Therefore, it is challenging to extract natural dyes in a fast, environmentally friendly, and low-cost way. The ultrasonic extraction method (UAE) uses the cavitation effect of ultrasonic waves to destroy plant cell structure, accelerate the solvent into the cell, speed up the dissolution of intracellular substances, and improve the yield, which can reduce the effect of temperature on the extract, and at the same time the application. It can reduce the effect of temperature on the extract and has a wide range of applications. However, too long a time will have specific effects, so it is necessary to control the appropriate time while the microwave extraction method (MAE) is mainly through the thermal effect of microwaves, the use of molecules that are susceptible to polarization, and the effect of ion conductivity to achieve direct heating of the material, with high efficiency, rapid, simple and more complete extraction of the characteristics (Kusuma and Mahfud 2017b; a, 2018).

Cinnamomum camphora (L.) wood, roots, branches, leaves, and fruits all have their uses. These materials have attracted the vision of researchers because of their renewable, green, versatile functions and excellent economy (Nguyen *et al.* 2018). Researchers have used natural dyes from camphor leaves and applied them in silk fabric dyeing to confer antimicrobial properties to fabrics or have used camphor wood chips as a biosorbent to remove alkaline dyes from aqueous solutions, thus treating waste solutions (Wang *et al.* 2014; Jiang *et al.* 2018), or used aqueous and methanolic extracts of fresh leaves of *C. camphora* fruit to achieve the inhibition of the growth of some algae (Chen *et al.* 2018). In the extraction method using natural colorants from *C. camphora* fruit leaves under the ultrasound-assisted mode, better extraction rates of chlorophylls and anthocyanins from fresh leaves were obtained. The extracted dyes were used to show excellent dyeing properties for both silk and wool, repeated use of the waste solution of natural dyes from *C. camphora* fruit leaves for coloring wool fabrics was achieved, and then the fabrics were tested for UV resistance (Gong *et al.* 2019a, 2019b; Zhou and Fu 2020; Feng *et al.* 2021; Mao *et al.* 2021; Rather *et al.* 2021). Camphor tree planting area is large, and *C. camphor* fruit production is prosperous; if not treated, substantial numbers of C. camphor fruit fall on the ground, which will cause environmental pollution and increase the municipal burden. Microwave extraction has the characteristics of high extraction efficiency, fast processing, and simplicity. Using microwave-assisted extraction of camphor fruit dye can reduce the extraction solvent, and improve the efficiency of dye extraction, to use camphor fruit more quickly and economically (Kusuma *et al.* 2018; Kusuma and Mahfud 2017a,b, 2018).

This study aimed to explore the extraction process of *C. camphora* fruit dyes and its performance using microwave-assisted extraction (MAE), which has the main advantages of high efficiency, fast temperature rise, short extraction time, low energy consumption, and low cost (Sarfarazi *et al.* 2020; Zuber *et al.* 2020). The extraction time is related to the cost and energy required. Because conventional extraction takes a long time and requires more cost and energy, microwave-assisted extraction consumes less energy (Kusuma *et al.* 2019; Kusuma and Mahfud 2017c).

Response surface methodology (RSM) combines mathematical and statistical methods successfully used to understand the effects of independent variables and optimize complex processes. The main advantages of RSM are that the method has fewer experimental points, high efficiency, and accurate predictions, while allowing the evaluation of independent variables and their interactions (Ansori *et al.* 2019; Iqbal *et al.* 2021; Kusuma *et al.* 2021, 2022). Box-Behnken design (BBD) is one of the many types of RSM. BBD optimizes the variables with a minimum number of experiments and helps to analyze the interactions between different variables further. In addition, it can avoid combinatorial treatments in extreme ranges and effectively estimate the factors of the quadratic model. The method has been widely used by researchers for experimental optimization. RSM was used to evaluate the microwave-assisted extraction of fruit dyes (Liu *et al.* 2011; Zhu *et al.* 2018; Li *et al.* 2020; Wu *et al.* 2020; Xiong *et al.* 2020; Zhao *et al.* 2020; Mao *et al.* 2022). The objective was to optimize the optimal conditions of the microwave-assisted extraction method of *C. camphora* fruit dyes using response surface methodology, while avoiding hazardous chemicals. The extraction time of camphor fruit dyes by microwave method is relatively fast, which is the advantage of the microwave method compared with the conventional method. The infrared spectral structure of *C. camphora* fruit dye was obtained by analysis. Its performance was tested under different pH values, different temperatures, and UV irradiation for the subsequent application of the *C. camphora* fruit dyes.

EXPERIMENTAL

Materials and Reagents

Cinnamomum camphora fruit peel powder (origin Nanjing Forestry University, moisture content 10%), pure water, sodium hydroxide, acetic acid, and qualitative filter paper were used in this study (Hangzhou, Zhejiang, China).

Apparatus and Equipment

A U-3900 type spectrophotometer (Hitachi High-Tech Science Corporation, Nako, Japan); P70F20CL-DG(B0) microwave oven (China Max. output power 700W; Galanz, Guangdong, China), OHRUS electronic balance (Sartorius, Beijing, China); Fy-1h-N vacuum pump (Zhejiang Feiyue; Wenling, China); LED UV lamp; HPLC- MS (Thermo Fisher Scientific, Massachusetts, USA); FD-1A-50 freeze dryer (Shanghai Biron Instrument Co., Ltd., Shanghai, China); and Fourier infrared spectrometer (Thermo Scientific, Waltham, MA, USA) were used in this study.

Cinnamomum camphora fruit pretreatment

The dyes in *C. camphora* fruit peel were distributed in the fruit peel's tissues and readily soluble in water; the fruit was first deseeded and dried. An appropriate amount of dried fruit peel was taken, the surface impurities were removed, and the powder was crushed with a powdering machine. The *C. camphora* fruit powder was obtained and sealed and stored for further use.

Dye extraction and absorbance determination

One gram of *C. camphora* fruit powder was weighed, and acetic acid was added to adjust the pH of pure water to 3 as the solvent. After microwave-assisted extraction, 1 mL of the extracted solution was taken in constant volume with the extraction solvent and diluted 10 times. U-3900 was used to measure the maximum absorption peak wavelength at 517 nm.

Single-factor test

Various material-liquid-ratios were set at 1:10, 1:20, 1:30, 1:40, and 1:50 g: mL; different microwave times as 20, 40, 60, 80, and 100 s; different microwave powers at 140, 280, 420, 560, and 700 W, were used in the examination of single factor changes. The other factors were controlled at a constant level, with the material-liquid-ratio kept at 1:30, microwave time kept at 40 s, and power kept at 280 W, to explore the effects of other factors on *C. camphora* fruit dyes.

Table 1. Single-factor Test: Factors and Levels

Level	Material-liquid Ratio (g:mL)	Microwave Time (s)	Microwave Power (W)
1	1:10	20	140
2	1:20	40	280
3	1:30	60	420
4	1:40	80	560
5	1:50	100	700

Cinnamomum camphora fruit dye response surface optimization

A test protocol was formulated using the Box-Behnken model using Design Expert 11 software (Stat-Ease, 11, Minneapolis, MN, USA). The test factors selected were microwave time (A), microwave power (B), and material-to-liquid ratio (C) based on the results of the single-factor test, and the absorbance was used as the response value to design a three-factor, three-level response surface analysis test for coding analysis and to obtain a quadratic multiple regression model. The central test was repeated three times. Factor levels were designed as shown in Tables 2 and 3.

Table 2. Box-Behnken Test Factors and Levels

Level	Factors		
	A: Microwave Time (s)	B: Microwave Power (W)	C: Material to Liquid Ratio (g:mL)
-1	60	280	1:10
0	80	420	1:15
1	100	560	1:30

Table 3. Response Surface Test Scheme and Results

Specimen Number	A Microwave Time (s)	B Microwave power (W)	C Material to Liquid Ratio (g:mL)	R (Abs)
1	0	0	0	0.457
2	0	1	-1	0.287
3	1	0	-1	0.274
4	0	0	0	0.489
5	0	-1	-1	0.189
6	-1	0	-1	0.253
7	-1	-1	0	0.277
8	0	0	0	0.509
9	0	0	0	0.465
10	0	-1	1	0.505
11	0	1	1	0.593
12	1	-1	0	0.343
13	-1	0	1	0.441
14	1	0	1	0.613
15	0	0	0	0.515
16	-1	1	0	0.38
17	1	1	0	0.577

RESULTS

Effect of Material-to-Liquid Ratio on *Cinnamomum camphora* Fruit Peel Dye

The absorbance of *C. camphora* fruit dye extract was influenced by the feed-liquid ratio (as in Fig. 1a). The dye absorbance reached a maximum at a feed-liquid ratio of 1:20 (g: mL). When the feed-liquid ratio decreases, the concentration gradient difference between the extraction solvent and the material is weak. The mass transfer driving force is low, which is not conducive to dye precipitation. When the feed-liquid ratio increases, the solubility of dye molecules increases. However, the excess solvent dilutes the saturated extraction solution as the feed-liquid ratio increases, thus decreasing the balsam fruit dye absorbance (Oberoi and Sogi 2017). Therefore, when conducting response surface optimization experiments, the material-liquid ratios of 1:10, 1:15, and 1:30 were selected as the three levels of response surface optimization factors.

Effect of Microwave-assisted Extraction Time on *Cinnamomum camphora* Fruit Dye

When the extraction time was 20 s to 80 s, the absorbance of *C. camphora* fruit dye extract increased with the extraction time (as in Fig. 2b), and the dye absorbance reached its maximum value at 80 s. When the extraction time was 100 s, the dye stability was destroyed due to the high temperature, which decreased absorbance (Sharmila *et al.* 2019). Therefore, extraction times of 60, 80, and 100 s were selected as triple levels when performing response surface optimization experiments.

Effect of Microwave Power on *Cinnamomum camphora* Fruit Dye Extraction

When the microwave power was varied within the range 140 to 420 W, the absorbance values of the *C. camphora* fruit dye solution changed as shown in Fig. 1c. The absorbance increased with increasing power. The absorbance value showed a decreasing trend when the microwave power continued to increase after 420 W, probably because the microwave power was too high to destroy the structure of the fruit dye. Therefore, 280, 420, and 560 W microwave powers were selected as three levels for response surface optimization experiments.

A comparison of influencing factors of dye extraction from *C. camphora* fruit dye by microwave-assisted extraction is shown in Fig. 2; solid-liquid ratio had the most significant influence on dye extraction, followed by microwave time and microwave power.

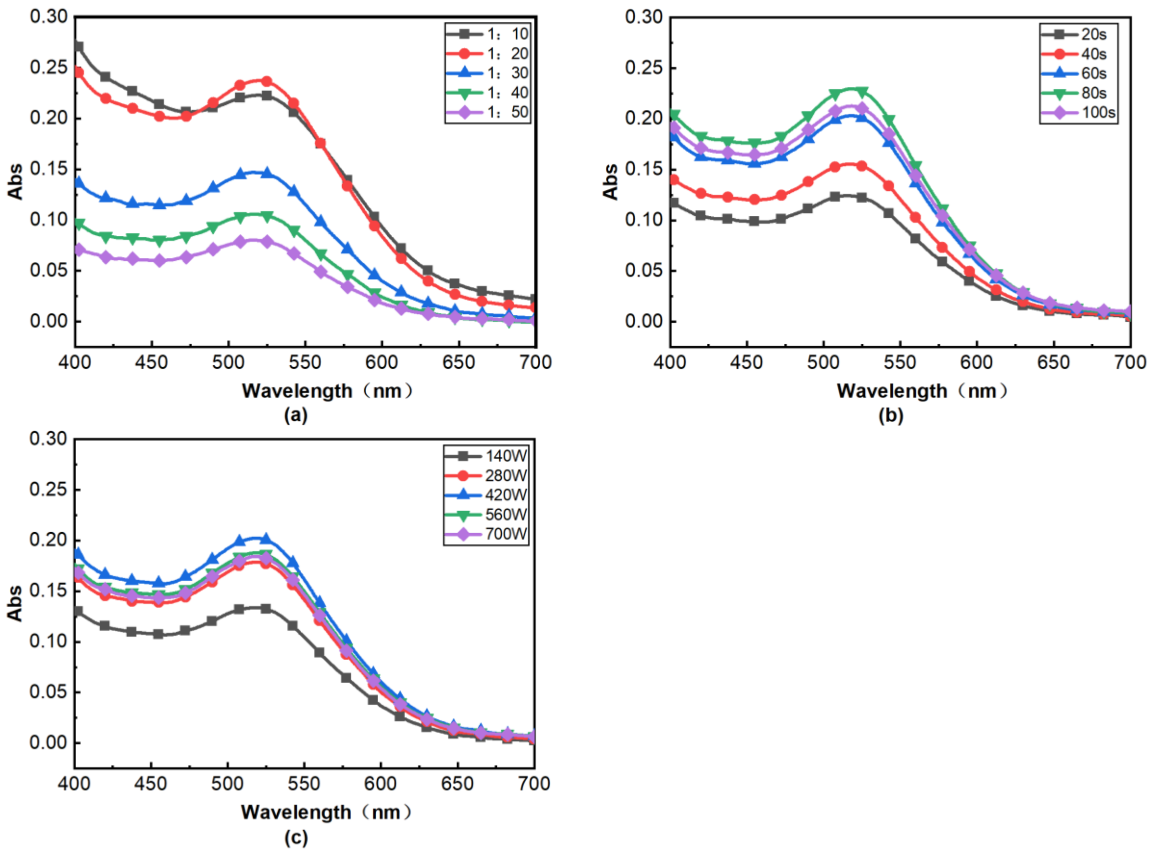

Fig. 1. (a)The influence of solid-liquid ratio on dye absorbance, (b)The influence of microwave time on dye absorbance, and (c)The influence of microwave power on dye absorbance

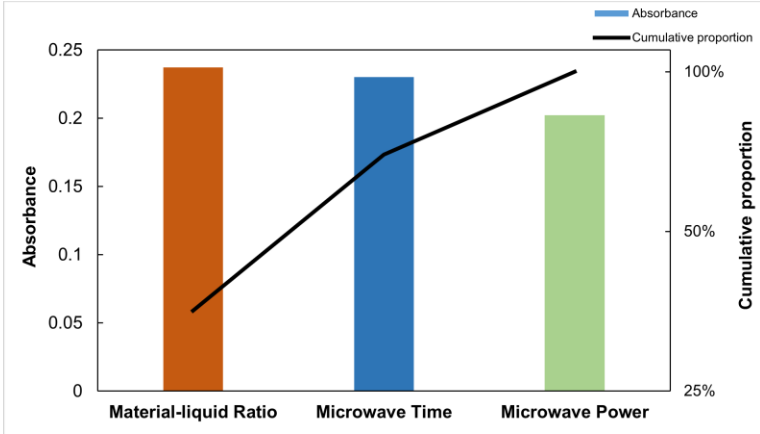

Fig. 2. Effects of material-liquid ratio, microwave time, and microwave power on absorbance of *C. camphora* fruit dyes

Results of Response Surface Optimization Design and Regression Modeling

As the absorbance comparison of the content of dyes in natural dyes is more intuitive, the absorbance was used as the response quantity to evaluate the camphor fruit dyes. Test factor level and response surface test design are shown in Table 2 and Table 3.

The regression models of the *C. camphora* fruit dye extraction process parameters were established according to the results of the response surface test (Table 3). The quadratic polynomial regression equations of the response values of the test evaluation index (based on the absorbance of *C. camphora* fruit dye extract) on the independent variables A (microwave time), B (microwave power), and C (material-to-liquid ratio) were obtained as follows,

$$\begin{aligned}&\text{Absorbance of } \textit{C. camphora} \text{ fruit dye (R)} = 0.487 + 0.0.057 \times A \\ &+ 0.0654 \times B + 0.1436 \times C + 0.0328 \times AB + 0.0378 \times AC \\ &- 0.0025 \times BC - 0.0455 \times A^2 - 0.0473 \times B^2 - 0.0463 \times C^2 \qquad (1)\end{aligned}$$

where A (microwave time), B (microwave power), and C (material-to-liquid ratio) are the test (input) variables.

Table 4. Statistical Analysis – Sequential Fitting of Model for *C. camphora* Fruit Dye Extraction

Sequential Model Sum of Squares						
Source	**Sum of Sq.**	**d f**	**Mean Square**	**F-Value**	**p-value**	
Mean vs Total	3.02	1	3.02			
Linear vs Mean	0.2252	3	0.0751	20.51	< 0.0001	
2FI vs Linear	0.0100	3	0.0033	0.8888	0.4798	
Quadratic vs 2FI	0.0303	3	0.0101	9.76	0.0068	Suggested
Cubic vs Quadratic	0.0046	3	0.0015	2.30	0.2186	Aliased
Residual	0.0027	4	0.0007			
Total	3.29	17	0.1938			
Lack of Fit Tests						

Source	Sum of Sq.	d f	Mean Square	F-Value	p-value	
Linear	0.0449	9	0.0050	7.52	0.0338	
2FI	0.0349	6	0.0058	8.76	0.0272	
Quadratic	0.0046	3	0.0015	2.30	0.2186	Suggested
Cubic	0.0000	0				Aliased
Pure Error	0.0027	4	0.0007			
Model Summary Statistics						
Source	**Std. Dev.**	**R^2**	**Adj.R^2**	**Pred.R^2**	**PRESS**	
Linear	0.0605	0.8256	0.7853	0.7199	0.0764	
2FI	0.0613	0.8623	0.7797	0.6460	0.0966	
Quadratic	0.0322	0.9734	0.9393	0.7155	0.0776	Suggested
Cubic	0.0258	0.9903	0.9611		*	Aliased

Sequential model fitting and ANOVA values are shown in Tables 4 and 5. The analysis of variance and the significance test of the model coefficients were performed on the model with the absorbance of the fruit dye as the evaluation index (as in Table 5), from which it can be concluded that the model was highly significant $p < 0.01$, ($p < 0.05$, $p < 0.01$), and the misfit term F was 0.2186, indicating that the misfit was not significant relative to the pure error. The coefficient of determination of the regression model was 0.9734, indicating that the model can explain 97.34% of the variation, the model fits well, the experimental error is within a reasonable range, and the optimization of the *C. camphora* fruit dye extraction process with this model can yield good results. The primary terms A, B, and C of the model were significant ($p < 0.05$, $p < 0.01$), and the primary term F value in the quadratic regression equation was used as the basis for comparison of the effect of each parameter on the absorbance of the fruit dye. It was found that the magnitude of the effect of the three factors on the absorbance of the fruit dye was C > B > A in order, *i.e.*, material-liquid ratio > microwave power > microwave time. The p-values of AB, AC, and BC were all greater than 0.05, indicating that the interaction among the three was not significant, and each factor had different effects on the absorbance of camphor fruit dye, so different extraction effects would be achieved by adjusting different factors. The effect of the interaction term followed the order AC > AB > BC. The R^2 values for quadratic and cubic terms were 0.97 and 0.99 (Kusuma *et al.* 2017; Kusuma and Mahfud 2017a,c), which reflects a high degree of validity of the model *C. camphora* fruit dye extraction (Table 4).

Table 5. Regression Model Variance Analysis

Source	Sum of Squares	d f	Mean Square	F-value	P-value	
Model	0.2655	9	0.0295	28.50	0.0001**	Significant
A	0.0260	1	0.0260	25.11	0.0015*	
B	0.0342	1	0.0342	33.03	0.0007**	
C	0.1650	1	0.1650	159.41	0.0000045**	
AB	0.0043	1	0.0043	4.14	0.0812	
AC	0.0057	1	0.0057	5.51	0.0513	
BC	0.0000	1	0.0000	0.0241	0.8809	
A^2	0.0087	1	0.0087	8.42	0.0229	
B^2	0.0094	1	0.0094	9.08	0.0196	
C^2	0.0090	1	0.0090	8.70	0.0214	
Residual	0.0072	7	0.0010			
Lack of Fit	0.0046	3	0.0015	2.30	0.2186	Not Significant
Pure Error	0.0027	4	0.0007			
Cor Total	0.2728	16				
R^2 0.9734						
* When $p < 0.05$, significant difference; ** When $p < 0.01$, high significant difference						

Analysis of Interaction Effects

The interaction of factors affecting the dye absorbance of camphor fruit can be reflected in the gradient change of the response surface (Figs. 3a, 3b). The slope of the response surface reflects the sensitivity of *C. camphora* dye when the treatment conditions change. The steep slope of the response surface indicates that the response value is sensitive when the treatment condition changes. The slope of the response surface is gentle, which means that the response value is not sensitive when the treatment condition changes.

According to the three-dimensional response surface diagram and two-dimensional contour figure, when any factor of solid-liquid ratio, microwave time, and microwave power is 0, the interaction of the other two factors can reflect the influence on the absorbance of *C. camphora* fruit dye. It can be found that the solid-liquid ratio has the most significant influence on the absorbance of *C. camphora* fruit dye. With the increase in solid-liquid ratio, microwave time, solid-liquid ratio, and microwave power, the absorbance of *C. camphora* dye also increases. However, when the ratio of solid to liquid, microwave time, and microwave power increase to a certain extent, the absorbance of *C. camphora* dye tends to decline. The elliptical shape of the contour diagram shows that the interaction between the two factors is significant. The circular contour map indicates that the interaction between the two factors is insignificant, and the test results are consistent with the model variance analysis in regression.

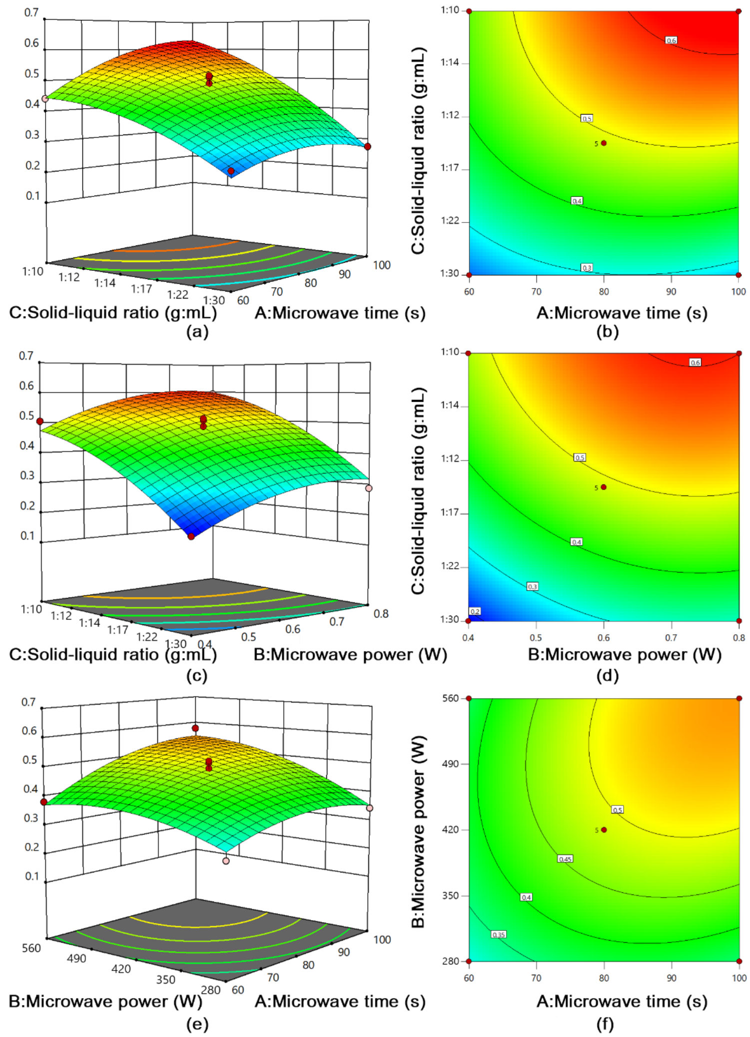

Fig. 3. Response surface plot (a) and contour plot (b) of the ratio and microwave time on the absorbance; Response surface plot (c) and contour plot (d) of the absorbance by the ratio of liquid to microwave power; Response surface plot (e) and contour plot (f) of microwave power *versus* microwave time on the absorbance during *Cinnamomum camphora* fruit dye extraction

Determination of Optimum Extraction Conditions of Dye from *Cinnamomum camphora* Fruit

The extraction process of *C. camphora* fruit bark can be obtained from the response surface analysis data: material-liquid ratio 1:19.61(g: mL), microwave time 88.722 s, and microwave power 420 W. At this time, the absorbance was 0.42. Considering the feasibility of laboratory operation, the extraction process conditions were modified: material-liquid ratio 1:20 (g: mL), microwave time 90 s, and microwave power 420 W.

After three parallel tests, the average absorbance was 0.44, similar to the simulated value of 0.42, indicating that the effect of response surface optimization of *C. camphora* fruit peel extraction conditions reached the predicted level.

In order to verify the applicability of the model equation for predicting the optimal out-of-condition response, the predicted value was 0.445 at the 70 s, 420 w, and 1:15 (g: mL), and the mean value was 0.45 after three parallel tests, which was similar to the simulated value, further demonstrating the validity of the model.

Compared with the 2h required by two-phase extraction (Wang *et al*. 2017), the time required by microwave-assisted extraction optimized by response surface method was significantly reduced, which demonstrates the advantage of microwave extraction method in the application of natural dyes. *C. camphora* fruit dye extracted under the optimal process obtained by response surface test can be applied to wood dyeing to reduce or replace the application of chemical dyes in wood dyeing. Under the premise of green and sustainable development, it can enrich the color system of wood decoration and improve the added value.

Infrared Spectrum Analysis of Dye in *Cinnamomum camphora* Fruit

Figure 4 shows the infrared spectra of *C. camphora* fruit dye, from which it can be seen that the fruit dyes have characteristic solid absorbance peaks at 3270, 2929, 1722, 1590, 1401, 1260, 1021, and 772 cm^{-1}. A broad absorbance peak near 3270 cm^{-1} shows O-H telescopic vibrations. The CH_2 telescopic vibrations peak is presented at 2929 cm^{-1}. Firm absorbance peaks are present around 1722 and 1590 cm^{-1}. Peaks found at 1401 and 1260 cm^{-1} correspond to the vibration of the C-O and C-O-C bonds. The absorbance peaks observed between 772 and 1200 cm^{-1} indicate that the molecule contains C-H out-of-plane deformation vibration (Table 6).

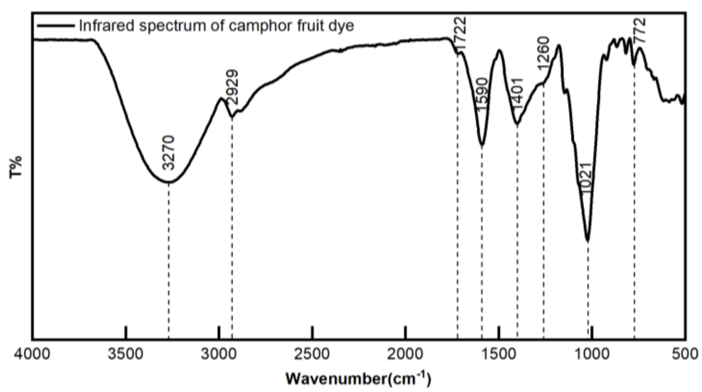

Fig. 4. Infrared spectrum of *Cinnamomum camphora* fruit dyes

Table 6. Characteristic FTIR Spectral Frequencies and Peak Attribution of the *Cinnamomum camphora* Fruit Dyes

Peak	Functional Group	Group Description
3270	O-H	O-H Telescopic vibrations
2929	CH_2	CH_2 Telescopic vibrations
1722	C=O	C=O Telescopic vibrations
1590	C-H	C-H Telescopic vibrations
1401	O-H	O-H Vibration of bending
1260	C-O-C	C-O-C Antisymmetric telescopic vibration
1021	C-O	C-O Telescopic vibrations
772	C-H	C-H Out-of-plane deformation vibration

HPLC-MS Analysis of *Cinnamomum camphora* Fruit Dyes

HPLC-MS is a qualitative analysis technique for natural products. The chromatogram of *C. camphora* fruit dye was analyzed by LTQ-Orbitrap. The mobile phase solution consisted of 5% acetonitrile and 95% formic acid with a concentration of 0.1%. The sample usage was 5 μL, and the positive ion mode electrospray mass spectrometry was used. The HPLC-MS spectra of *C. camphora* fruit dye is shown in Fig. 5, and the liquid mass spectra is of molecular weight 449 and 595.The mass-to-charge ratio (m/z) of 449 was detected at 14.42 min, 15.49 min, and 13.44 min. *Centaurea* diglycosides with a mass/charge ratio (m/z) of 595 were detected at 14.18 min and 14.18 min, respectively. The results are consistent with the conclusions of many researchers (Fig. 6).

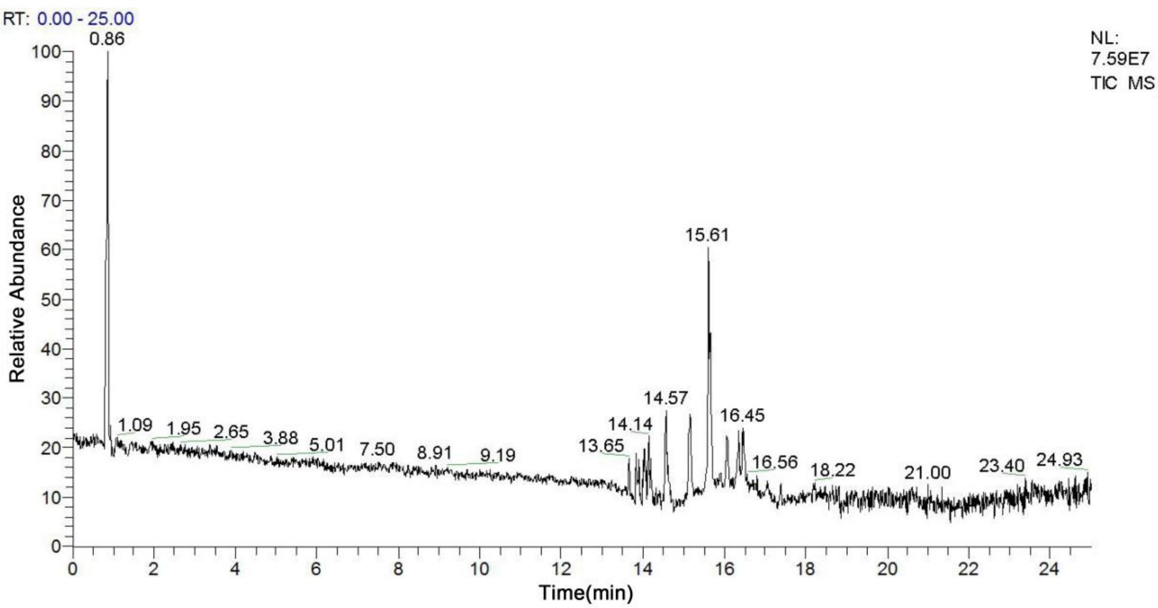

Fig. 5. HPLC-MS spectra of *Cinnamomum camphora* fruit dyes

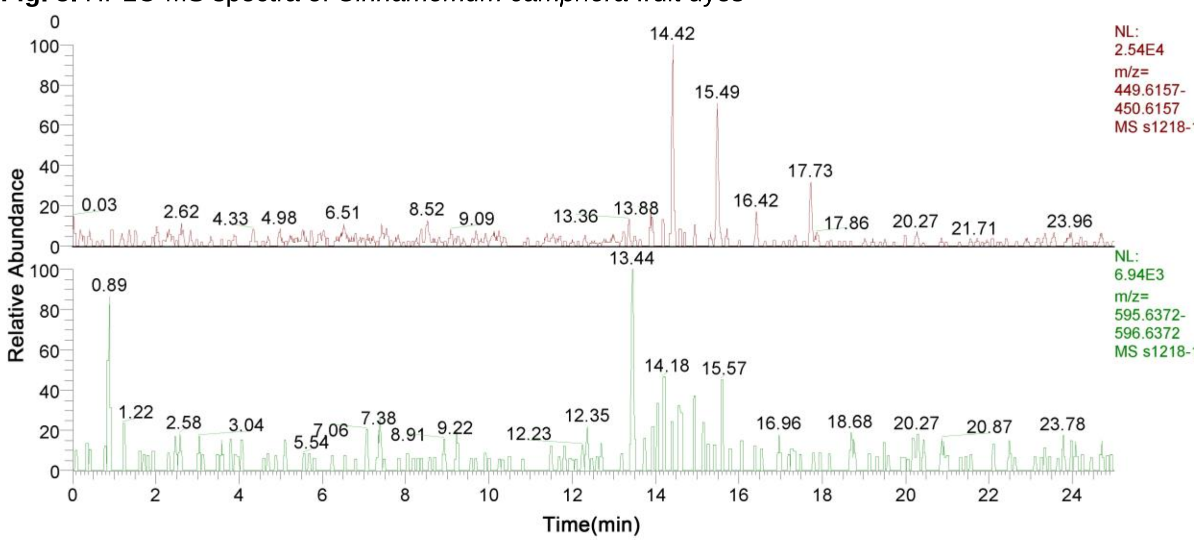

Fig. 6. HPLC-MS profile at molecular weight 449 and 595

Stability Study of *Cinnamomum camphora* Fruit Dyes

Effect of pH on the stability of Cinnamomum camphora fruit dye

Figure 7a shows digital images of the *C. camphora* fruit dye solution after 2 h at different pH values; the pH values of the dye solution from left to right are 2, 4, 6, 8, 10, and 12 in order. The appearance changes of the fruit dye solution at pH 2, 4, and 6 are not apparent; as the pH value was increased, the fruit color dye solution became dark green. As can be seen from Fig. 4d, when the pH was at 8, 10, and 12, the absorbance value appeared to be significantly reduced in the UV spectra. The morphology changed considerably, probably because the strong alkali caused some damage to the fruit dye's molecular structure, which decreased its absorbance value. Therefore, it can be concluded that the *C. camphora* fruit dyes are more stable under acidic environments.

Effect of temperature on the stability of Cinnamomum camphora fruit dye

Figure 7b shows the digital images of the fruit dye solution after 2 h of heating in a water bath at different temperatures, from left to right, the temperature of the dye solution is at 40 °C, 50 °C, 60 °C, 70 °C, and 80 °C. The color of the dye was slightly changed. The naked eye weakens the color of the dye solution. It can be seen from Fig. 4e that the temperature had a certain degree of influence on the stability of camphor fruit dye. The absorbance value decreased relatively fast from 40 °C to 50 °C and 50 °C to 80 °C. After 80 °C, its absorbance value showed a decreasing trend. It can be seen that camphor fruit dyes can be affected by temperature, leading to a decrease in absorbance, so the high temperature should be avoided in the process of extraction, use, and storage of *C. camphora* fruit dyes.

Effect of UV light on the stability of Cinnamomum camphora fruit dye

Figure 7c indicates the digital images of *C. camphora* fruit dye solution after different times of UV irradiation, the staining from left to right in the order of 1, 2, 3, 4, and 5 h of UV irradiation. It can be seen from Fig. 4f that with the extension of UV irradiation time, the absorbance value of the fruit dye decreased under UV irradiation. This observation is because the fruit dye’s degradation is accelerated by UV irradiation; thus, UV can reduce the stability of *C. camphora* fruit dye. Hence, when producing, storing, and transporting this dye, it is beneficial to avoid UV irradiation as much as possible.

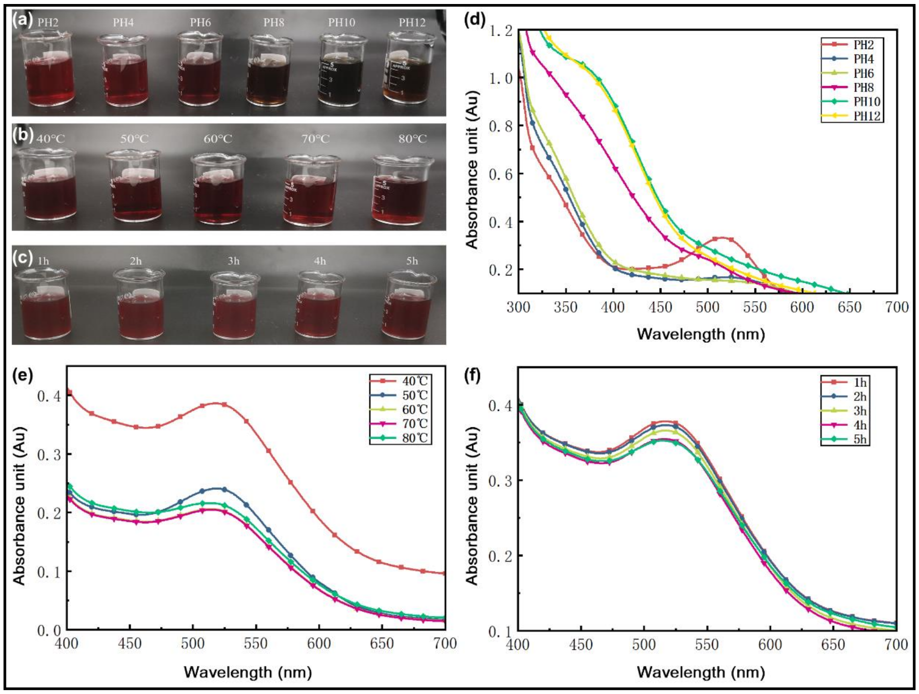

Fig. 7. Digital images of the effect of (a) pH, (b) temperature, and (c) UV irradiation on *Cinnamomum camphora* fruit dye; (d) UV spectra of the effect of pH on the absorbance, (e) UV spectra of the effect of temperature on the absorbance, and (f) UV spectra of the effect of UV irradiation on the absorbance of *Cinnamomum camphora* fruit dye

Environmental impacts of Microwave-assisted extraction of *Cinnamomum camphora* fruit dye

Table 7 summarizes the environmental impacts of microwave-assisted extraction of *C. camphora* fruit dye on electricity consumption and carbon dioxide emissions. Regarding power consumption (0.42 kW h versus 1.5 kW h) and carbon dioxide release (330g versus 1178 g), microwave-assisted extraction has a lower environmental impact than conventional solvent heating extraction. These data suggest that microwave-assisted extraction has great potential to replace traditional extraction methods (Kusuma and Mahfud 2017c; Kusuma *et al*. 2019).

Table 7. Energy and Environmental Impacts

	Microwave-Assisted	**Conventional Extraction**
Extraction time (min)	1.5	120
Electric consumption (kW h)	0.42	1.5
RA_{CO2} (g)	330	1178

CONCLUSIONS

1. The dyes in *Cinnamomum camphora* fruit were extracted by microwave extraction, and the influencing factors of dye extraction were optimized by response surface methodology based on single-factor tests (Liu *et al.* 2020). The optimized process conditions of *C. camphora* fruit dye extraction was 1:20 (g/mL) material-to-liquid ratio, microwave time of 90 s, and microwave power of 420 W. Under this condition, the absorbance of dye in the fruit extract was 0.44, which reached the predicted level. It indicates that the microwave-assisted extraction process of the fruit is accurate and reliable. The test was stable and reliable, which provided a reference for the study of green extraction of *C. camphora* fruit (Wang and Zhou 2023).
2. Compared with the traditional method, the optimized microwave-assisted extraction process can shorten the dye extraction time, increase extraction efficiency, reduce unnecessary resource consumption, and improve production efficiency when applied to the actual production process.
3. Based on the results of the response surface test to analyze the infrared spectrum of the C. camphor fruit dye, The structure of *C. camphor* fruit dye contains O-H, CH_2, C=O, C-O-C, C-H bond, and other functional groups. HPLC-MS results revealed that in *C. camphor* fruit dye, the mass-to-charge ratio (m/z) of 449 was detected at 14.42 min and 15.49 min, 13.44 min. *Centaurea* diglycosides with a mass/charge ratio (m/z) of 595 was detected at 14.18 min.
4. The stability of *C. camphor* fruit dye was tested, and the results showed that the stability of the fruit dye decreased to a certain extent under high-temperature conditions, and it is suitable for storage or use in low to medium temperature environment; pH had a more significant influence on the dye solution, and under the acid or alkaline conditions dyestuff was easily changed. The dye was shown to be strongly degraded by extended UV irradiation, so it should be avoided during storage in long time light irradiation. The environmental impact of microwave-assisted extraction of camphor fruit dyes indicates that microwave-assisted extraction has excellent potential.

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