Application of Full Factorial Design to Optimize Solvent Free Microwave Extraction of Eucalyptus Citriodora Essential Oil

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Abstract— The solvent free microwave extraction (SFME) of essential oil from eucalyptus was optimized using a full factorial design in terms of oil yield to determine the optimum extraction conditions. Several experiments were carried out with varying parameters like, extraction time, microwave power and type of sample for two level of each. The mathematical model applied form a first order regression equation. The predicted values calculated by the regression model were in good agreement values. The results showed that the extraction time is most prominent factor followed by microwave power level and sample type for extraction process. An average of 0.97 % of eucalyptus oil can be extracted using existing setup. The optimum condition for the eucalyptus oil extraction using SFME, were the extraction time 30 minutes, microwave power level 640 watt and sample type cuts sample. SFME proves a green and promising technique for essential oil extraction.

Key words: SFME, Eucalyptus essential oil, modeling desirability function, optimization

I. INTRODUCTION

The principal aim of green and chemistry and engineering is to reduce chemical related impact on human health and to search alternative environmentally friendly and energy efficient production methods. Green and clean extraction method which can offer the more natural products, free from toxic solvents. The search of such green extraction methods were highly emphasized in essential oils industries since last decade because of consumer's preference towards natural products. Essential oil which are volatile extract of the species, medical and aromatic plants. The history of essential oil extraction and their use for various purposes from is as old as mankind.

Essential oils are volatile, natural base products, which are found in spices, aromatic and medicinal plants. The Extraction of essential oils is well known from old ages when pure essential oil and crude extract of essential oil bearing plants, herbs and grasses were in use for various medicinal and fragrances, flavors, preservatives and insect repellents purposes. [1-2]

Eucalyptus is a diverse genus of flowering tree and shrubs including a distinct group with a multiple-stem melee growth habit in myrtle family (myrtaceae), its botanical name is eucalyptus citriodora. There are more than 700 species of eucalyptus, mostly native to Australia, and a very small number are found in adjacent areas of new Guinea and Indonesia. Species of eucalyptus are cultivated throughout the tropics and subtropics including the America, Europe, Africa, Middle East, China and Indian subcontinent. [3] Its essential oil is sweet and the primary source of cooling sensation and has achieved a high economic value. [4] Essential oil is find main application as flavor in chewing gums, toothpastes, confectionary, tobacco and alcoholic beverage. [5] The eucalyptus citriodora have also found applicable in shampoo, and skin care product. It is commonly used to soothe or treat nausa, vomiting, abdominal pain, indigestion, irritable bowel and blotting. It is also used in aroma therapy. [6-7] In India 30 to 40 tons of Essential oil is extracted from eucalyptus per year. [8]

Steam distillation is the primary method to extract the eucalyptus essential oil for commercial product. [9] Many other methods can be used for extraction of essential oils from eucalyptus, e.g. hydro-distillation (HD), Supercritical fluid extraction, solvent extraction, soxhlet, supercritical water extraction and direct thermal desorption. [10-11] However these essential oil compounds are thermally sensitive and vulnerable to chemical changes. These extraction methods may result in losses of some volatile compounds, low extraction efficiency, long extraction time, degradation of unsaturated or ester compounds through thermal or hydrolytic effects and toxic solvent residue in the extract. [12] To overcome these problems some new "green" techniques in essential oil extraction have been developed by researchers. These techniques typically use less solvent and energy, such as ultrasound and microwave.

Microwave assisted process is a recent technique for essential oil extraction from plant materials, patented by federal department of environment, Canada. [13] Today, MAP is well known to selective and volumetric heating of target, less extraction time, high product quality. This technology finds application from analytical laboratory systems to industrial extractor. [14]

In this study, solvent free microwave extraction (SFME) was adopted for oil extraction of eucalyptus citriodora. The extraction process was optimized in terms of essential oil yield using full factorial design, determining those variables that most influenced essential oil yield. A 2^3 full factorial plan was carried out to model process. The simple first degree model was used which gave a representation of the response function according to the variables.

II. MATERIALS AND METHODS

Eucalyptus leaves were collected from a private farm at Sehore. The authenticity of variety was verified by department of botany, SSSUTMS University, Sehore. Plants were cleaned and dried in a dark room at 25°C. Solvent free microwave extraction (SFME) was performed at atmospheric pressure with a microwave frequency of 2450 MHz using a household microwave oven; which was mechanically modified to perform the hydro distillation, as shown in figure 1. This was a multimode microwave reactor with a maximum delivered output power of 800 W and input power of 1200W, having the voltage supply of 230 volt and dimensions of the oven cavity was 206mm (H) x 300mm (W) x 302mm (D).

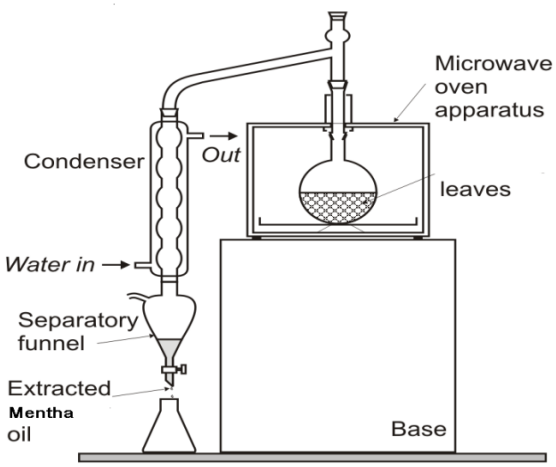

Fig. 1: Microwave extraction apparatus.

A. *Extraction of Essential Oil:*

The eucalyptus leaves were properly cleaned and chopped in to 1-2 cm. long pieces. Before extraction of essential oil, plant was stored in shade with limited air circulation and for three different drying durations. The moisture lost during 24 hour was 9-10 %. A 100 gm of sample was placed in reactor with the runs were taken at three different levels of time and microwave power. Light yellow colored oil, with a lemon like odor, was obtained which was separated and dried over the minimum amount of anhydrous sodium sulfate to remove traces of moisture. The percentage oil yield is expressed as follows;

$$\text{Oil yield } (\%) = \frac{\text{mass of extracted oil}}{\text{mass of sample}} \times 100$$

B. *Essential Oil Chemical Composition:*

A standard gas liquid chromatography was used for the analysis of essential oil of eucalyptus obtained from SFME. The analysis was carried out at fragrance and flavors development centre (FFDC), Kannauj, Uttar Pradesh (India). GLC analysis of the eucalyptus oil was performed on slip mode HP (Hewlett – Packard, make HP-5890) having detector flame ionization with a carbowax column (30 m x .25m i.d., film thickness .325 µm). The oven temperature was programmed from 50 ^{0}C (initial time 5 min) to 230 ^{0}C with 50 ^{0}C/min and injector have the temperature 230 ^{0}C and 240 ^{0}C respectively. The identification of some compound was done by comparing with the retention times of variables standard. Area percentage in the chromatogram was used to knowing the percentage of each compound in the oil.

C. *Mathematical Treatment:*

To investigate the efficiency of extraction process on eucalyptus essential oil yield, a two level, 2^3 full factorial designs was constructed. Three variables were chosen namely; extraction time (X_1), microwave power (X_2); sample size (X_3). Each independent variable had 2 levels which were coded as -1 and +1. The coded values of independent variables were found from equation;

$$\text{Xi} = \frac{Xi - Xo}{\Delta x}$$

Where x_0 the base is value at the center of experiment domain and x_i is original variables and Δx is the average value of the difference between highest and lowest values. The eight run in design matrix of 2^3 full factorial designs are set up by randomization. A multiple regression, first degree model was used to express the response as a function of all three factors; which are centered and reduced variables.

$$Y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{22} x_2 x_3 + \beta_{123} x_1 x_2 x_3$$

Where average β_0, main effect β_i and interaction β_{ij} and β_{ijk}. Design exert version 8.0.7.1 (trail version) was applied for performing the experimental design and the data analysis.

III. RESULT AND DISCUSSION

A. *Statistical Analysis:*

Table 2 shows the of design matrix of experimental outcome, as carried out at above specified levels as soon in table 1 and expressed as extraction yield (Y) of essential oil extracted from eucalyptus sample it is observed from response table that an interaction model best represent the microwave extraction process in terms of coded variables;

$$Y = .31 + .062\ X_1 + .050\ X_2 + .050\ X_3 + .041\ X_1 X_2 + .031\ X_2 X_3 - .029\ X_1 X_3 + .021\ X_2 X_3$$

This regression equation shows that the optimum yield should be located in the experimental domain or much closed to it, as the values of β's are not very high. An average yield of essential oil we can extract from the current experimental set up is $\beta_0 = 0.31$ % for the current levels of the factors.

Levels	X1 (min)	X2 (watt)	X3 (size in cm)
Basic level (0)	20	464	1
High level (+1)	30	640	1.5
Low level (-1)	10	288	1
Interval	10	176	1

Table 1: Coded and natural variables in 2^3 factorial design.

	Code variables			Oil yield
Run	x1	x2	x3	%
1	-1	-1	-1	0.2
2	1	-1	-1	0.26
3	-1	1	-1	0.18
4	1	1	-1	0.39
5	-1	-1	1	0.24
6	1	-1	1	0.22
7	-1	1	1	0.32
8	1	1	1	0.46
9	-1	-1	-1	0.21
10	1	-1	-1	0.26
11	-1	1	-1	0.18
12	1	1	-1	0.39
13	-1	-1	1	0.24
14	1	-1	1	0.22
15	-1	1	1	0.32
16	1	1	1	0.46

Table 2: 2^3 factorial design including the corresponding responses

To assess the goodness of the fit of the empirical model and to check the adequacy of model that has been generated by the factorial experiment, analysis of variance (ANOVA) was conducted at 95% confidence level (P=0.05) given in table 3.

Source	Sum of Squares	Degree of freedom	Mean square	F value	P value prob > f
Model	0.19	7	0.034	98.2	< .0001
A extraction time	0.067	1	0.051	210.7	<.0001
B power	0.052	1	0.062	190.8	< .0001
C sample	0.032	1	0.028	108.7	<.0001
AB	0.21	1	0.21	65.7	< .0001
AC	0.19	1	0.19	67.9	<.0001
BC	0.22	1	0.22	76.8	<0.0001
ABC	5.83E-03	1	5.83E-03		
Pure error	2.000E-0.003	8	2.000E-0.003		
Cor total	0.19	12			

Table 3: Analysis of variance

Result showed adjusted – R^2 (0.9783), an adequately high degree of correlation between experimental and predicted values. Coefficient of determination R^2 (0.9885) was desirably high (closed to 1), adequate precision (AP) 30.411, standard deviation (SD) 0.0.016, and coefficient of variation (CV) 0.6.32 %.Based on the ANOVA result, it is clear that the model is good simulation of degradation experiment. The predicted R^2 value is also in agreement with the adjusted R^2 value. Figure -2 shows the normal probability plot of the residuals for percentage of oil yield.

IV. CONCLUSIONS

Solvent free microwave extraction (SFME) was used for the extraction of useful essential oil from eucalyptus Citriodora. This technique is well known to its significant effects on essential oil bearing herbs and plants materials, application of this method result in a more natural aroma of essential with less extraction time in comparison to conventional methods like steam or water distillation. The extracted oil is free from residue of toxic solvents. It can be concluded from this study that duration of time is most dominant factor followed by microwave power and type of sample. For eucalyptus citriodora, solvent free microwave extraction proved a promising technique with high quality of essential oil.

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