Optimisation of *Croton gratissimus* Oil Extraction by *n*-Hexane and Ethyl Acetate Using Response Surface Methodology

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Abstract: The extraction of oil from *Croton gratissimus* seeds was studied using the three-factor five-level full-factorial central composite rotatable design (CCRD) of the response surface methodology (RSM). The effect of the three factors selected, viz., extraction time, extraction temperature and solvent-to-feed ratio on the extraction oil yield was investigated when *n*-hexane and ethyl acetate were used as extraction solvents. The coefficients of determination ($R^2$) of the models developed were 0.98 for *n*-hexane extraction and 0.97 for ethyl acetate extraction. These results demonstrated that the models developed adequately represented the processes they described. From the optimized model, maximum extraction yield obtained from *n*-hexane and ethyl acetate extraction were 23.88% and 23.25%, respectively. In both cases the extraction temperature and solvent-to-feed ratio were 35°C and 5 mL/g, respectively. In *n*-hexane extraction the maximum conditions were reached only after 6 min whereas in ethyl acetate extraction it took 20 min to get the maximum extraction oil yield. Oil extraction of *Croton gratissimus* seeds, in this work, favoured the use of *n*-hexane as an extraction solvent as it offered higher oil yields at low temperatures and reduced residence times.

Key words: *Croton gratissimus*, oil extraction, Central Composite Rotatable Design (CCRD), Response Surface Methodology (RSM), optimisation

1 Introduction

*Croton gratissimus* is a terrestrial plant of a semi-deciduous tree species belonging to the family of *Euphorbiaceae*. It is commonly found in rocky or stony terrains throughout much of the warmer and drier regions, from north-eastwards regions of South Africa to the horn of Africa³. Almost the entire plant (from roots to leaves) is used in traditional medicine to treat a wide variety of ailments. Recently it has been reported that oil extracted from *Croton gratissimus* seeds could be a promising feedstock in the large-scale production of biodiesel⁴. However, information that would help to effectively design and optimise industrial scale processes for *Croton gratissimus* oil extraction is not available in the literature. This motivated the present study, aimed at generating experimental oil extraction data in view of further valorisation of this African crop.

Generally, large-scale seed oil extraction is achieved either by mechanically pressing the seeds against a solid surface or by using chemical solvents (in hot water extraction, Soxhlet extraction and ultrasonic techniques). Alternatives to these well-established industrial processes include supercritical fluid extraction (SFE), ultrasonic-assisted extraction (UAE) and enzymatic oil extraction which are currently investigated by many researchers over the world⁵. The most common and efficient solid-liquid extraction method used in producing oil for biodiesel production is the solvent extraction method; where a solute fraction (oil) is transferred from a solid material (seed) to a liquid solvent⁶.

Sánchez-Arraolea et al.⁷ performed a Soxhlet extraction of *Jatropha curcas* using *n*-hexane, ethyl acetate and ethyl ether as extraction solvents. Of the three solvents used, the best yield was obtained with ethyl acetate extraction (54.3%). Extractions using *n*-hexane and ethyl ether gave oil yields of 47.7% and 45.9%, respectively⁸. In their study on biodiesel production from *Croton grassimus* oil, Bahadur et al.⁹ reported only one extraction data point, *i.e.*
23.5% oil yield when \( n \)-hexane was used as solvent. In the present study however, systematic yield measurements were undertaken at various conditions, with two different solvents, \( n \)-hexane and ethyl acetate.

In addition to experimental data, modelling studies are an important step towards industrial implementation of a process. The two major approaches towards modelling oil extraction from seeds, grains and fruits include theoretical and empirical models. The most commonly used theoretical correlations are the single stage washing and diffusion model, the two stage washing and diffusion model, the first order rate equation, the second order rate equation and the simplified model based on Ficks’s equation. These are kinetic models as they relate yield to contact time. In relation to thermodynamic modeling, enthalpy and entropy changes are generally determined from experimental data through the van’t Hoff equation. Popular empirical models for oil extraction are those based on response surface methodology and artificial neural network.

In this article, the effects of extraction time, extraction temperature and solvent-to-feed ratio on the oil extraction yield (measured in terms of mass of oil produced per mass of seeds used) of the two solvents, \( n \)-hexane (non-polar solvent) and ethyl acetate (polar solvent), in the extraction of oil from Croton gratissimus seeds are reported. Furthermore, response surface methodology coupled with the central composite design of experiments is used to model and optimise the extraction process in both cases. It is worth noting that \( n \)-hexane is currently the oil extraction solvent of choice in industry. However the rising concern over its environmentally-unfriendly nature justifies the quest for alternative solvents among the so-called generally recognised as safe solvents. One such solvent is ethyl acetate whose extraction ability is compared with that of \( n \)-hexane in this study. The effect of particle size on the extraction oil yield has been extensively studied by a number of researchers who showed that a decrease in the particle size results in an increase in the extraction rate leading to high extraction oil yields. It is for this reason that no attempts were made in this study to investigate the effect of particle size.

2 Materials and Methods

2.1 Materials

Croton gratissimus grains were obtained from selected trees in the city of Lubumbashi, Upper Katanga province, South-Eastern region of the Democratic Republic of the Congo (DRC). Grains were allowed to dry in open air under direct sunlight for 2 days. Thereafter they were de-shelled with a hammer to remove the 3-lobed fruit capsules and expose the seeds. A high speed kitchen blender was used to crush the seeds to the desired particle size. Classifica-

2.2 Experimental set up and extraction procedure

2.2.1 Experimental set up

A 100 mL jacketed beaker fitted with a reflux condenser was used for the extraction of oil. Agitation was achieved through the use of magnetic stirrer equipped with a speed controller. A rectangular hot water bath with an immersion temperature regulator-circulator was used to offer consistent heating at predetermined temperatures. Solvents were recovered from the extracted oil by heating the mixture in a round-bottomed flask fitted with a reflux condenser and immersed in a thermostated water bath.

2.2.2 Extraction procedure

A known mass of dried seeds (10 g) was placed in a 100 mL jacketed beaker fitted with a reflux condenser. For every gram of dried seeds fed to the beaker, a range of known volumes of extraction solvent was added (1.65 mL to 5.85 mL). A variable speed magnetic stirrer set at 600 RPM (revolutions per minute) was used for agitation. Different operating temperatures were explored and the minimum and maximum being 26.5°C and 68.5°C, respectively. Extraction times per batch ranged between 1.75 and 24.75 minutes. Uncertainties in measuring experimental parameters were estimated as ±0.5°C, 0.05 mL and 0.005 g for temperature, volume and mass, respectively. After the set time had elapsed and extraction completed, the miscella was filtered using a laboratory filtration equipment set up, i.e., a Buchner flask fitted with a dropping funnel and connected onto a vacuum supply line. To avoid solvent loss during vacuum filtration, another condenser was fitted to the dropping funnel. A mixture of oil and solvent was collected in the flask and the residue discarded. The mixture was then fed to a flash distillation equipment set up to separate the solvents from oil. The extraction yield was calculated as the ratio of the mass of the extracted oil to that of the Croton gratissimus seeds used in the experiment.

3 Experimental Design

The response surface methodology (RSM), used in this
Optimisation of Croton gratissimus Oil Extraction by n-Hexane and Ethyl Acetate

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Experiments were performed in a randomized order to minimize the effects of the uncontrolled factors. The Design-Expert® version 10 software was used to design the experiments and for regression and graphical analyses of the data obtained. The values of the yield were taken as the responses of the designed experiments. Statistical analysis of the model was performed to evaluate the analysis of variance (ANOVA)\(^{10}\).

The selection of levels (uncoded variables) for each factor was based on the literature related to the uses of n-hexane and ethyl acetate as solvents in liquid extraction operations\(^ {11}\). The lower levels of temperature were limited by mass transfer constraints. At low temperatures, the viscosity of the extract (oil) is at its highest and its diffusivity greatly reduced leading to low extraction rates\(^ {12}\). The upper levels of the temperature were limited by boiling temperatures of the selected solvents. Higher temperatures are beneficial to the solubility of Croton gratissimus oil in the extracting solvent, and could accelerate the extraction process. However, increasing temperature will bring about not only the increase in costs in view of industrialisation but also lipids oxidation\(^ {13}\).

The extraction yield (\(Y\)) response was used to develop an empirical model that correlated the response to the oil extraction variables using the second-order polynomial equation:\(^ {14}\)

\[
Y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ij} X_i^2 + \sum \sum \beta_{ijk} X_i X_j + \epsilon
\]

where \(Y\) is the predicted response, \(\beta_0\) is the constant coefficient, \(\beta_i\) is the linear coefficient, \(\beta_{ij}\) is the interaction coefficient, \(\beta_{ijk}\) is the quadratic coefficient, \(X_i\) and \(X_j\) are the uncoded values of the independent variables, ET, TT and SF, and \(\epsilon\) is the random error\(^ {14}\).

### 4 Results and Discussion

The RSM, utilising the CCD and ANOVA, was applied in the optimization of selected independent variables to obtain maximum oil extraction yields. The effects of extraction time, extraction temperature and solvent-to-feed ratio on the oil yield were determined for n-hexane and ethyl acetate and the results are summarised in Table 2.

From the experimental runs, the highest oil yield of 23.92% was obtained when n-hexane was used as a solvent in the extraction of oil from Croton gratissimus seeds.

#### 4.1 n-Hexane Extraction

When n-hexane was used as an extraction solvent in different combinations of independent variables, the average yield was found to be 18.92% from a range of between 5.24% and 23.92%, as shown in Table 2. Results obtained in this work in the temperature range from 47.5°C to 60°C are close to the only oil yield data point reported by Bahadur et al.\(^ {5}\) when they carried out an n-hexane oil extraction on croton gratissimus seeds for 6 hours in a round-bottomed flask fitted with a mechanical stirrer at a temperature of 50°C and a solvent-to-feed ratio of 2.5 mL/g. They found an extraction oil yield of 23.5% from Croton gratissimus seeds of an average particle size of 1500 ± 50 μm. It is worth mentioning that the present study is more comprehensive than that of Bahadur et al.\(^ {5}\) who had a rather different aim from systematically investigating the extraction process. The present study involves the use of two solvents, i.e., ethyl acetate and n-hexane. Attempts were made to model and optimise the process whereas Bahadur et al.\(^ {5}\) merely reported a single oil extraction experiment using n-hexane only.

#### 4.1.1 Modelling and Optimisation

The experimental data obtained in the n-hexane oil ex-

| Variables | Symbols | Levels |
|-----------|---------|--------|
| Extraction time (min) | ET | -α | -1 | 0 | +1 | +α |
| Extraction temperature (°C) | TT | 1.25 | 5 | 12.5 | 20 | 24.75 |
| Solvent-to-feed ratio (mL/g) | SF | 26.5 | 35 | 47.5 | 60 | 68.5 |
|             |        | 1.65 | 2.5 | 3.75 | 5 | 5.85 |
traction was analyzed by the ANOVA and the following quadratic regression model that best describes this extraction process was obtained:

\[ Y_{\text{H}} = -5.633 - 0.087X_1 - 0.314X_2 + 14.316X_3 + 0.004X_1X_2 - 0.017X_1X_3 - 0.006X_2X_3 - 0.001X_1^2 + 0.003X_2^2 - 1.339X_3^2 \tag{2} \]

where \( Y_{\text{H}} \) is the percentage oil extracted using \( n \)-hexane, \( X_1, X_2 \) and \( X_3 \) are the uncoded values of the independent variables, viz., extraction time \((ET)\), extraction temperature \((TT)\) and solvent-to-feed ratio \((SF)\), respectively.

The analysis of variance (ANOVA) for the experimental results of the CCD is shown in Table 3a. From the developed quadratic model, the Fisher’s \( F \)-test gave an \( F \) value for the model of 62.75 with a very low probability value \((p < 0.0001)\). This implied that the model was significant and that a value that large had only a 0.01% chance to be due to noise. The goodness of fit of the model was evaluated by the determination coefficient \((R^2)\), adjusted determination

| Experimental run | Coded independent variable levels | % Extraction Yield |
|------------------|----------------------------------|--------------------|
|                  | Extraction time (min) | Extraction temperature (°C) | Solvent-to-feed ratio (mL/g) | Type of factor | \( n \)-Hexane \((Y_{\text{H}})\) | Ethyl acetate \((Y_A)\) |
| 1                | -1 | -1 | -1 | Factorial | 15.03 | 13.66 |
| 2                | 1  | -1 | 0  | Factorial | 13.51 | 12.67 |
| 3                | -1 | 1  | -1 | Factorial | 13.67 | 14.66 |
| 4                | 1  | 1  | -1 | Factorial | 14.24 | 14.03 |
| 5                | -1 | -1 | 1  | Factorial | 23.92 | 23.01 |
| 6                | 1  | -1 | 1  | Factorial | 22.46 | 22.98 |
| 7                | -1 | 1  | 1  | Factorial | 22.88 | 22.09 |
| 8                | 1  | 1  | 1  | Factorial | 22.13 | 21.02 |
| 9                | -1.682 | 0 | 0 | Axial | 18.79 | 18.28 |
| 10               | 1.682 | 0 | 0 | Axial | 20.54 | 21.41 |
| 11               | 0   | -1.682 | 0 | Axial | 21.07 | 18.93 |
| 12               | 0   | 1.682 | 0 | Axial | 21.42 | 20.39 |
| 13               | 0   | 0   | -1.682 | Axial | 5.24  | 5.28  |
| 14               | 0   | 0   | 1.682 | Axial | 22.83 | 20.36 |
| 15               | 0   | 0   | 0   | Centre | 20.17 | 20.46 |
| 16               | 0   | 0   | 0   | Centre | 19.66 | 20.32 |
| 17               | 0   | 0   | 0   | Centre | 20.41 | 20.89 |
| 18               | 0   | 0   | 0   | Centre | 20.35 | 20.32 |
| 19               | 0   | 0   | 0   | Centre | 20.23 | 18.69 |
| 20               | 0   | 0   | 0   | Centre | 19.92 | 20.44 |

Table 3a \( n \)-Hexane extraction: Analysis of variance (ANOVA) for the quadratic model.

| Source of variation | Sum of squares | Degrees of freedom | Mean square | \( F \)-value | \( p \)-value | \( \text{Prob > } F \) |
|---------------------|---------------|--------------------|-------------|---------------|-------------|-----------------------------|
| Model               | 375.76        | 9                  | 41.75       | 62.75         | < 0.0001    |                             |
| Residual            | 6.65          | 10                 | 0.67        |               |             |                             |
| Lack of fit         | 6.25          | 5                  | 1.25        | 15.50         | 0.00459     |                             |
| Pure error          | 0.40          | 5                  | 0.081       |               |             |                             |
| Total               | 382.41        | 19                 |             |               |             |                             |

\[ CV = 4.31\% \quad R^2 = 0.98 \quad \text{Pred. } R^2 = 0.88 \quad \text{Adj. } R^2 = 0.97 \quad \text{Adeq. Precision} = 30.526 \]
The coefficient of determination ($R^2$) of the model was 0.9826, indicating that the model adequately represented the real relationship between the parameters chosen (Fig. 1). This meant that only 0.17% of the total variations was not explained by the developed model. The coefficient of variation (C.V.) of 4.31% indicated that the model was reproducible. The predicted $R^2$ of 0.8775 was in reasonable agreement with the adjusted $R^2$ of 0.9669, i.e., the difference was less than 0.2. The Adequate Precision, a measure of the signal-to-noise ratio that should ideally be greater than 4, was also evaluated and found to be high enough at 30.526. This meant that the developed model had a high degree of precision and could thus be used to navigate the design space.

The regression equation (equation 2) showed a negative correlation with extraction time ($X_1$) and extraction temperature ($X_2$) but was positively correlated with the solvent-to-feed ratio ($X_3$). This implied that an increase in either the extraction time or the extraction temperature resulted in a decreased extraction oil yield whereas an increase in solvent-to-oil ratio increased the extraction oil yield. This trend of oil yield change as a function of solvent-to-feed ratio was consistent with mass transfer principles. In mass transfer the concentration gradient between the solid and the bulk of the liquid is the main driving force; and this driving force is greater when higher solvent-to-feed ratios are used. A larger magnitude of the coefficient for solvent-to-feed ratio ($p < 0.01$) when compared to the magnitudes of the coefficients of the other two independent variables ($p > 0.05$) further proved that solvent-to-feed ratio had a greater significance in $n$-hexane oil extraction process.

4.1.2 Effect of independent variables on percentage extraction oil yield using $n$-hexane

Figure 2 shows the response surface plots of extraction oil yield, for various combinations of extraction time, solvent-to-feed ratio and extraction temperature, when $n$-hexane was used as an extraction solvent. Figure 2a reveals that oil extraction yield is a very weak function of extraction temperature and time. As evidence, an almost flat plot was observed showing a 1.08% reduction in the extraction oil yield when the extraction temperatures and the extraction times were increased from 35°C to 60°C and from 5 min to 20 min, respectively. In both these cases the solvent-to-feed ratio was kept constant at 5 mL/g. On the other hand, the solvent-to-feed ratio had a significant effect on the percentage oil yield. The percentage oil yield showed a markedly sharp increase, from 6.79% to 23.78%, when the solvent-to-feed ratio was increased from 1.65 mL/g to 5 mL/g and the extraction temperature kept constant at 35°C (Fig. 2b). Figure 2c showed an increase in the percentage oil yield from 6.69% to 23.77% over the full range of solvent-to-feed ratio values (1.65 mL/g – 5 mL/g) when the extraction time was kept constant at 6 min.

From the developed model (equation 2), it was found that an optimum extraction oil yield of 23.88% could be obtained under the following operating conditions: extraction time, 6 min; extraction temperature, 35°C and solvent-to-feed ratio, 5 mL/g.

4.2 Ethyl Acetate Extraction

When ethyl acetate was used as an extraction solvent in different combinations of independent variables, the extraction oil yield obtained ranged between 5.28% and
23.01% with an average of 18.50% (Table 2).

4.2.1 Modelling and Optimisation

The experimental data obtained in the ethyl acetate oil extraction was analysed by the ANOVA and the following quadratic regression model that best describes this extraction process was obtained:

\[ Y_E = -23.513 + 0.011X_1 + 0.195X_2 + 17.138X_3 - 0.001X_1X_2 + 0.007X_1X_3 - 0.042X_2X_3 + 0.002X_1^2 - 0.0002X_2^2 - 0.0001X_3^2 \]

where \( Y_E \) is the percentage oil extracted using ethyl acetate, \( X_1, X_2 \) and \( X_3 \) are uncoded values of the independent variables, viz., extraction time (ET), extraction temperature (TT), and solvent-to-feed ratio (SF), respectively.

Equation 3 showed all linear coefficient terms as positive meaning that they all had a positive effect on the percentage oil yield. An increase in the extraction time, extraction temperature or the solvent-to-feed ratio resulted in an increase in the percentage oil extraction yield. But larger magnitudes of the coefficient of solvent-to-feed ratio \( (p < 0.01) \) when compared to the magnitudes of the coefficients of the other two independent variables \( (p > 0.05) \) indicated that the solvent-to-feed ratio was the most significant independent variable in controlling the ethyl acetate oil extraction process.

The developed quadratic model was evaluated through the analysis of variance (ANOVA) and the results are shown in Table 3b. The ANOVA of the regression model (Equation 3) showed that the quadratic model was highly significant. This was evident from the Fisher’s \( F \)-test which gave an \( F \) value for the model of 34.60 with a very low probability value \( (p < 0.0001) \). The goodness of fit of the model was evaluated by the determination coefficient \( (R^2) \), adjusted determination coefficient \( (\text{Adj. } R^2) \) and the coefficient of variance \( (C.V.) \). The value of the coefficient of determination \( (R^2) \) was found to be 0.9689, indicating that only 0.31% of the total variations was not explained by the developed regression model (Fig. 3). The predicted \( R^2 \) of 0.8282 was in reasonable agreement with the adjusted \( R^2 \) of 0.9409.

### Table 3b Ethyl Acetate extraction: Analysis of variance (ANOVA) for the quadratic model.

| Source of variation | Sum of squares | Degrees of freedom | Mean square | \( F \)-value | \( p \)-value |
|---------------------|----------------|--------------------|-------------|---------------|--------------|
| Model               | 351.00         | 9                  | 39.00       | 34.60         | < 0.0001     |
| Residual            | 11.27          | 10                 | 1.12        |               |              |
| Lack of fit         | 8.36           | 5                  | 1.67        | 2.87          | 0.13565     |
| Pure error          | 2.91           | 5                  | 0.58        |               |              |
| Total               | 362.27         | 19                 |             |               |              |

\( CV = 5.74\% \quad R^2 = 0.97 \quad \text{Pred. } R^2 = 0.83 \quad \text{Adj. } R^2 = 0.94 \quad \text{Adeq. Precision} = 22.696 \)
The Lack of fit $F$-value of 2.87 implied that the Lack of Fit was not significant and that there was a 13.57% chance that a Lack of Fit that large could occur due to noise. The Adequate Precision was also evaluated and found to be high enough at 22.696. This meant that the developed model had a high degree of precision. The coefficient of variation (C.V.) of 5.74% indicated that the model was reproducible.

4.2.2 Effect of independent variables on percentage extraction oil yield using ethyl acetate

The effects of extraction temperature, extraction time and solvent-to-feed ratio on the percentage extraction oil yield when ethyl acetate was used as an extraction solvent are shown in the surface plots in Fig. 4. The profile of the surface plot in Fig. 4a, where the solvent-to-feed ratio was kept constant at 5 mL/g, was almost flat, indicating a slight increase in the extraction oil yield when the extraction temperatures and the extraction times were varied within their low and high factor levels. The $p$ values of these two factors were far greater than 0.05 (0.6056 and 0.6328, respectively), indicating that their effects on the extraction oil yield were insignificant within the range of factor levels chosen. Figure 4b shows the extraction yield as a function of solvent-to-feed ratio and extraction time when the extraction temperature is at 35°C. An increase in the extraction oil yield was observed, from 4.63% to 22.92%, when the solvent-to-feed ratio was increased from 1.65 mL/g to 5 mL/g. The same could not be said for the extraction time that showed a mere 0.61% increase in the extraction oil yield when the extraction time was varied between 5 min and 20 min. Figure 4c showed an equally similar increase in the percentage oil yield from 4.66% to 22.90% over the full range of solvent-to-feed ratio values (1.65 mL/g – 5 mL/g) when the extraction time was kept constant at 20 min.

From the developed model, it was found that an optimum extraction oil yield of 23.25% could be obtained under the following operating conditions: extraction time, 20 min; extraction temperature, 35°C and solvent-to-feed ratio, 5 mL/g.
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