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Modeling and optimization of *Terminalia catappa* L. kernel oil extraction using response surface methodology and artificial neural network

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ABSTRACT

In this study, response surface methodology (RSM) and artificial neural network (ANN) were used to optimize *Terminalia catappa* L. kernel oil (TCKO) yield. Solvent extraction method was used for the oil extraction, with n-hexane as the extracting solvent. The highest oil yield was obtained at 55 °C, 150 min, and 0.5 mm. The physicochemical properties of the TCKO were determined using standard methods. Gas chromatographic (GC) analysis and Fourier Transform Infrared (FTIR) were respectively, used to determine the fatty acid composition and prevalent functional groups in TCKO. At optimum conditions of temperature, particle size and extraction time, the RSM predicted oil yield was 62.92%, which was validated as 60.34%, whereas ANN predicted yield was 60.39%, which was validated as 60.40%. The results of the physicochemical characterization of TCKO showed that the dielectric strength (DS), viscosity, flash and pour points values were 30.61 KV, 20.29 $mm^2\ s^{-1}$, 260 °C, and 3 °C, respectively. Physicochemical characterization and FTIR results of TCKO indicated its potential industrial application, especially as transformer fluid. Fatty acids compositions result indicated that the oil was highly unsaturated; while XRD results of *Terminalia catappa* L. kernel (TCK) samples obtained, both before and after extraction, showed difference in their peaks and corresponding intensities, due to the damage effect of solvent. Finally, the obtained optimization results indicated that ANN was a better and more effective tool than RSM, due to its higher R^2 and lower RMS values.

1. Introduction

Vegetable oil is one of the essential substances that are obtained from seeds and nuts. It often determines the applicability of the seeds/nuts for both local and industrial uses. Recently, the demands for more seeds and nuts with high oil yield have increased. This is attributed to the need for the production of bio based oils for industrial applications. The high biodegradability of bio based vegetable oils has increased their demand, hence encouraging the commercial cultivation of such seeds and nuts. As a result of this, more improved and high oil yield varieties of seeds and nuts have been developed in many agricultural research institutes across the globe (Leakey et al., 2003). Consequently, over the years, these improved varieties have been massively planted and have grown into important economic trees, bearing these oil seeds and nuts in large quantities (Manyong et al., 2005). Some of such oil seeds and nuts include but not limited to soya bean, palm trees, *Jatropha curcas*, groundnut, *Terminalia catappa* L., *Irvingia gabonensis* (IG) etc.

Lately, global concern about the safety of petroleum and synthetic based oils, as well as the environmental concern associated with their non-biodegradability nature, have led to more attention being shifted towards the used of vegetable based oils for both domestic and industrial applications. That notwithstanding, the choice of the seeds/nuts is also of great importance. This is to ensure that more of the non-food competing ones are used for industrial purposes. Hence, one of the reasons for the choice of *Terminalia catappa* L. in this present work, since it is non-food competing.

In Nigeria, *Terminalia catappa* L. is mostly planted for the provision of shades and for ornamental purposes, with little or no attention paid to the seeds and kernels. As such, it is highly underutilized for both domestic and industrial purposes, except for the minimal consumption of the fleshy fruit and kernels by little children in some rural areas (Agu, 2014). *Terminalia catappa* L. belongs to combretaceae family and is native to Malaya and the Andamanso (Iha et al., 2014). The oil content of the kernels varies and ranges between 49 and 64% (Menkiti et al., 2015; Iha et al., 2014; Dos Santos et al., 2008). Its kernel is rich in protein and carbohydrate, with the percentage ranges from 17% to 18.39% and 25.61%, respectively (Adepoju et al., 2014). *Terminalia catappa* L. kernel (TCK) contains hung quantity of fatty acid with the total saturated fatty

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acid content reported to be 34.2%, while the total unsaturated fatty as 64.5% (Iha et al., 2014).

With relatively high oil yield of *Terminalia catappa* L. kernel, there is need to develop new methods of oil extraction from it, as well as carry out the optimization and statistical analysis of the extraction process parameters. Like other seeds/nuts, *Terminalia catappa* kernel oil can be extracted by a number of methods, such as supercritical method, ultrasound-assisted method, mechanical process and solvent extraction methods (Da Porto et al., 2012; Yolmeh et al., 2014; Li et al., 2009; Sulaiman et al., 2013). However, higher yield and low turbid oil, as well as relatively easy operation of solvent extraction equipment, encourage its utilization, when compared to the other methods mentioned. Thus, reason behind its application in the present study using n-hexane as the extracting solvent.

Solvent extraction method has been utilized in the past for the extraction of vital oil from a number of seeds and nuts, such as *Jetropha curcas* (Devappa et al., 2010), soyabean (Navarro Cerutti et al., 2012), castor seed (Pradhan et al., 2012), coconut (Sulaiman et al., 2013), tobacco seed (*Nicotiana tabacum* L.) (Stanisavljević et al., 2007), grape seed (Fernandez et al., 2010), Avocado (*Persea americana*) (Dos Santos et al., 2014), *Colocynthis vugaris Shrad* seed (Agu et al., 2018) and hemp seed (Kostić et al., 2013), because of its efficiency and relatively inexpensive nature. The extraction yield of oil obtained from *Terminalia catappa* L. by solvent extraction method using soxhlet extractor and n-hexane as solvent is mainly influenced by the extraction process parameters. The effects of process parameters like extraction time, particle size and temperature were often evaluated for modeling and optimization purposes. Hence, the modeling, optimization and statistical evaluation of the process parameters and response (oil yield), during oil extraction process, using response surface methodology (RSM) and artificial neural network (ANN), become very important.

Response Surface Methodology (RSM) is a statistical and mathematical tool used for process optimization that involves the responses that are to be optimized, based on the process parameters (Betiku and Ajala, 2014). It is a vital optimization tool that is used in a number of engineering applications (Nazghelichi et al., 2011). The usefulness of RSM in industrial research has been seen especially for situations whereby the numbers of process variables influencing the response(s) are many (Betiku and Ajala, 2014; Gopinath et al., 2010). One of the very important advantages of RSM is the reduction in the number of experimental runs, and still has the capability of providing acceptable results (Onoji et al., 2017; Betiku and Ajala, 2014). RSM also has the advantage of generating second-order polynomial equation, which relates the dependent(s) or response(s) to the independent or process parameters (Onoji et al., 2017). RSM has been used for the optimization of oil extraction yields. For instance, Onoji et al. (2017), optimized *Hevea brasiliensis* (rubber seed) oil extraction using RSM and obtained 42.98 wt% oil yield at optimum conditions. Similarly, Reshad et al. (2015) and Kostić et al. (2013), respectively used RSM to optimize oil extraction yields of rubber seeds and hampseed, with obtained yields of 49.36 wt% and 30.43 wt%, respectively, at their different optimum conditions. In the work of Agu et al. (2015), RSM was successfully used to optimize *Terminalia catappa* kernel oil yield, with yield of 60.45 wt% at optimum conditions.

Artificial neural networks (ANNs) are the most common artificial learning tool (Betiku et al., 2015; Gueguim Kana et al., 2012). ANNs works on the basis of information processing that is synonymous to the human brain. As such, it consists of clusters of interconnected neurons, hence, can function as a modeling tool for solving complex nonlinear processes between the input and output of a system (Ameer et al., 2017; Nazghelichi et al., 2011). Aside its modeling ability, ANNs can also be used for prediction, control and optimization of processes (Fayyazi et al., 2015; Waewsak et al., 2010; Esonye et al., 2019a). Its optimization ability has been seen in the optimization of bioprocesses, such as biogas production (Gueguim Kana et al., 2012; Waewsak et al., 2010), methyl ester production (Esonye et al., 2019b), as well as in the esterification and transesterification processes (Ofoefule et al., 2019). Over the years, ANNs have been applied in solving several engineering, science, medicine, mathematics, neurology, metrology, psychology and biology problems (Betiku and Ajala, 2014). However, in terms of oil extraction modeling and optimization, a number of works have successfully used ANNs. For instance, Onoji et al. (2017) successfully modeled and optimized *Hevea brasiliensis* (rubber seed) oil extraction yield using ANNs. Similarly, Zahedi and Azarpour (2011) optimized passiflora seed oil extraction using ANNs. ANNs were also successfully used by Shokri et al. (2011) and Kostić et al. (2013) for the modeling and optimization of *Pimpinella anisum* L. seed and hampseed oils extraction, respectively. However, to the best knowledge of the authors, there is no literature information that compares the efficacies of RSM and ANNs in the modeling of *Terminalia catappa* L. kernel oil extraction.

Therefore, this work focused on the influence of these process parameters on the oil yield of TCK, as well as the modeling of solvent extraction of *Terminalia catappa* L. kernel oil using RSM and ANNs, for possible industrial application of the base fluid as transformer oil. Furthermore, the physicochemical properties of the oil were examined using standard methods. Furthermore, gas chromatography (GC) and Fourier Transform Infrared (FTIR) were used to determine the fatty acid compositions and functional groups present in the oil, respectively. Finally, the X-ray diffractometry was used to reveal the characteristics of the crystalline structure of both the TCK and the residue left after extraction.

2. Materials and methods

2.1. Materials

TCK were collected from Nsukka Local Government Area, Enugu State, Nigeria. N-hexane and other chemicals of analytical grades were procured from Conraws laboratory chemical vendor in Enugu. The reagents were used without further purification.

2.2. Methods

2.2.1. Sample preparation

Prior to oil extraction from *Terminalia catappa* L. kernels (TCK), its external coatings were carefully cracked, and the kernels were removed and cleaned. The kernels were sun dried, and thereafter oven dried at 60 °C for 12 h. The dried TCK were then ground using electrical grinder. Different sieve plates sizes were used to separate the ground TCK in other obtain 5 different particle size diameters of 0.5, 1.0, 1.5, 2.0 and 2.5 mm.

2.2.2. Terminalia catappa L. kernel (TCK) oil extraction

Solvent extraction method using soxhlet extractor was used for the extraction process. The process was carried out following the procedure described by the authors in their previous works on TCK oil extraction (Menkiti et al., 2015; Agu et al., 2015). The oil yields obtained at the end of the extraction process under the specified different process conditions (see Menkiti et al., 2015; Agu et al., 2015) were calculated and recorded. The solute to solvent ratio used for the process was 1:10 (15 g, 150 ml), mass-by-volume. The entire process was carried out three times and the average values that were determined were then reported. The solvent extraction using n-hexane, as well as the determination of the percentage oil yields at different conditions, were carried out according to the Association of Official Analytical Chemists AOAC (1979) method no. 963.15.

The TCK oil yield was calculated using Eq. (1).

$$\%Oil\ yield = \frac{weight\ of\ oil\ extracted\ (g)}{weight\ of\ sample\ (g)} \times 100. \quad (1)$$

2.2.3. Physicochemical properties of Terminalia catappa L. kernel oil (TCKO)

Physicochemical characterizations were carried out on the TCKO sample. Standard methods were used for the determination of these vital properties. The oil density (AOAC 985.19), iodine value (AOAC 993.20) and acidity/acid value (AOAC 969.17), were determined according to AOAC (1990) approved techniques. On the other hand, the viscosity and dielectric strength were determined following ASTM D445 (2011) and IEC 60156 (2003) standard methods, respectively. Furthermore, pour and flash points were determined using ASTM D97 and ASTM D93, standard test method, respectively (ASTM, 2008). The flash point was measured using a Pensky-Martens closed cup tester. The moisture content was determined using ASTM E203 standard test method. Every physicochemical property was measured three times, and the average values were calculated and recorded.

2.2.4. Fatty acid composition determination of TCKO

The fatty acid profile of TCKO was analyzed using gas chromatography (GC) (Shimadzu GC–14B, Model 910), according to AOAC (1990) 996.06. This equipment was equipped with a flame ionization detection system. The column was HP 88 capillary column (0.25 mm i.d. × 100 m, film thickness 0.25 μm – Shimadzu Corporation, Tokyo, Japan). The oven temperature program and other operating conditions are as described in the author's previous work on GC analysis of TCKO (Menkiti et al., 2017). Each fatty acid quantity acid was determined from the percentage area of the individual fatty acid (Menkiti et al., 2017; Zahedi and Azarpour, 2011). The analysis was carried out three times.

2.2.5. Fourier Transform Infrared Spectroscopy (FT-IR) analysis

The FTIR analysis of the TCKO sample was carried out using BUCK Scientific Infrared Spectrophotometer Model 530.

2.2.6. X-ray diffraction (XRD)

The XRD analyses of the *Terminalia catappa* L. kernels (TCK) and its residue after oil extraction were carried out using EMPYREAN analytical, Netherlands.

2.3. Design of experimental for TCKO extraction and statistical analysis using RSM

Oil extractions from each of TCK sample using n-hexane was modeled and optimized using central composite design (CCD). A three-factor-five-level CCD, which generated 20 experimental runs that were afterward carried out, was used in the modeling and optimization studies. The independent/process variables considered for the optimization process include temperature X_1 (°C), particle size X_2 (mm), and extraction time X_3 (min). The temperature range of 35 to 55 °C, particle size of 0.5 to 2.5 mm, and extraction times of 30 to 150 min were carefully chosen. The coded and uncoded levels of these independent variables are shown in Table 1. The experimental design was based on CCD, and by using the five levels, the CCD generated 20 experimental runs as shown in Table 2. Table 2 shows the run order, variable conditions and the columns for the experimental, predicted and residual values of the TCK oil yields. Multiple regression analysis was used as a tool for the assessment of the effects of the independent variables (temperature, particle size and extraction time) on the dependent variable (% oil Yield) (Boonmee et al., 2010). Second-order polynomial model, shown in Eq. (2) was used to analyze the response surface regression procedure for predicting the response variable (%Y).

$$\%Y = \beta_0 + \beta_1 X_1 - \beta_2 X_2 + \beta_3 X_3 + \beta 11 X \tag{2}$$

where %Y is the predicted response (% oil yield), β_0 is the intercept term, β_1, β_2, and β_3 are the linear coefficients, β_{11}, β_{22}, and β_{33} are the quadratic coefficients, β_{12}, β_{13}, and β_{23} are the interactive coefficients, and X_1, X_2, and X_3 are the coded independent variables. MINITAB 17.0 software was used in this study for the regression analysis and analysis of variance (ANOVA). Confidence levels of 95% were the basis for test of statistical significance.

2.4. Modeling and optimization by ANN

Commercial ANN software, using Neural Network Toolbox of MATLAB R2017a software package was used in this study. The TCKO yield was successfully predicted using Multilayer Full Feedforward (MFFF) and Multilayer Normal Feedforward (MNFF) neural networks. The networks were trained by Incremental Back Propagation (IBP). The ANN architecture consisted of an input layer with three neurons, an output layer with one neuron and a hidden layer. The optimal network topology was determined with one hidden layer, while the number of neurons in hidden layer and the hidden layer transfer function and output layer were determined iteratively, through the development of series of networks. The individual ANNs were trained using 1000 iterations. The CCD experimental data were used for both training and testing.

2.5. Verification of data

The predicted TCKO oil yields obtained from RSM and ANN were compared with the experimentally obtained responses. This was aimed at the evaluation of the efficacy of the both optimization methods. Coefficient of determination (R^2) and root mean square (RMS) were determined and used in the identification of the best ANN models architecture. Similarly, the RMS was used for comparing the RSM and ANN predicted and experimental values, for purpose of

Table 1
Uncoded and coded levels of independent variables of the TCKO extraction process.

Variable	Symbol	Level				
		Axial (−α)	Low	Center	High	Axial (+α)
		−1.633	−1	0	1	1.633
Temperature (°C)	X_1	28.67	35	45	55	61.33
Particle size (mm)	X_2	0.365	0.5	1.5	2.5	3.133
Time (min)	X_3	22.02	30	90	150	187.98

Table 2
CCD matrix of factors and responses of TCK oil yield by RSM.

Run	X_1	X_2	X_3	Oil Yield (%)		
				Actual	Predicted	Residual
1	−1.000	1.000	−1.000	24.80	21.60	3.20
2	1.000	−1.000	−1.000	34.90	35.48	−0.58
3	0.000	0.000	0.000	39.50	39.97	−0.47
4	1.000	1.000	1.000	42.00	42.43	−0.43
5	−1.000	−1.000	1.000	53.98	55.96	−1.98
6	1.000	−1.000	1.000	60.45	62.92	−2.47
7	0.000	0.000	0.000	40.00	39.97	0.03
8	0.000	0.000	0.000	39.95	39.97	−0.02
9	1.000	1.000	−1.000	28.99	26.27	2.72
10	−1.000	1.000	1.000	37.90	36.58	1.32
11	−1.000	−1.000	−1.000	30.85	29.69	1.16
12	0.000	0.000	0.000	40.11	39.97	0.14
13	−1.633	0.000	0.000	36.85	38.77	−1.92
14	0.000	−1.633	0.000	53.89	51.17	2.72
15	0.000	1.633	0.000	24.01	27.83	−3.82
16	0.000	0.000	1.633	53.80	51.27	2.53
17	0.000	0.000	0.000	40.35	39.97	0.38
18	1.633	0.000	0.000	49.08	48.27	0.81
19	0.000	0.000	0.000	40.25	39.97	0.28
20	0.000	0.000	−1.633	13.00	16.63	−3.63

determining the better optimization tool. Eqs. (3) and (4) were used to calculated the (R^2) and root mean square (RMS) values.

$$R^2 = 1-\sum_{i=1}^{n}\left(\frac{\left(x_{i,cal}-x_{i,\ exp}\right)^2}{\left(x_{avg,\ exp}-x_{i,\ exp}\right)^2}\right) \tag{3}$$

$$RMS = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\frac{x_{i,\ exp}-x_{i,cal}}{x_{i,\ exp}}\right)^2} \tag{4}$$

where n is the number of experimental data, $x_{i,\ cal}$ is the calculated values, $x_{i,\ exp}$ is the experimental values, and $x_{avg,\ exp}$ is the average experimental values.

Models accuracy is verified by the evaluation of the both the coefficient of determination (R^2) and root mean square (RMS) values. For a model to be highly acceptable, the R^2 should be as close as possible to 1, while the RMS values should be as small as possible (Agu et al., 2018).

2.6. Process flow chart

Fig. 1 shows the flow chart of the processes/steps that are contained in this work. From the chart, it could be seen that prior to the extraction of oil from *Terminalia catappa* L. kernel (TCK), the TCK sample was first prepared. This was done by sorting, drying and milling the TCK sample in other to obtain the required particles size. Design of experiment was done using central composite design (CCD). A three-factor-five-level CCD, which generated 20 experimental runs that were afterward carried out, was used in the modeling and optimization studies. Thereafter, oil was extracted from the TCK sample using solvent extraction method, and n-hexane as the extracting solvent. X-ray diffraction (XRD) analyses were carried out on the milled TCK and residue samples left after the extraction. Similarly, physicochemical characterization, Gas chromatographic (GC) analysis and Fourier Transform Infrared (FTIR) analysis, were carried out on the extracted TCK oil (TCKO). Modeling and optimization studies were carried out using response surface methodology (RSM) and Artificial Neural Network (ANN). Finally, verification/validation of the obtained data was then carried.

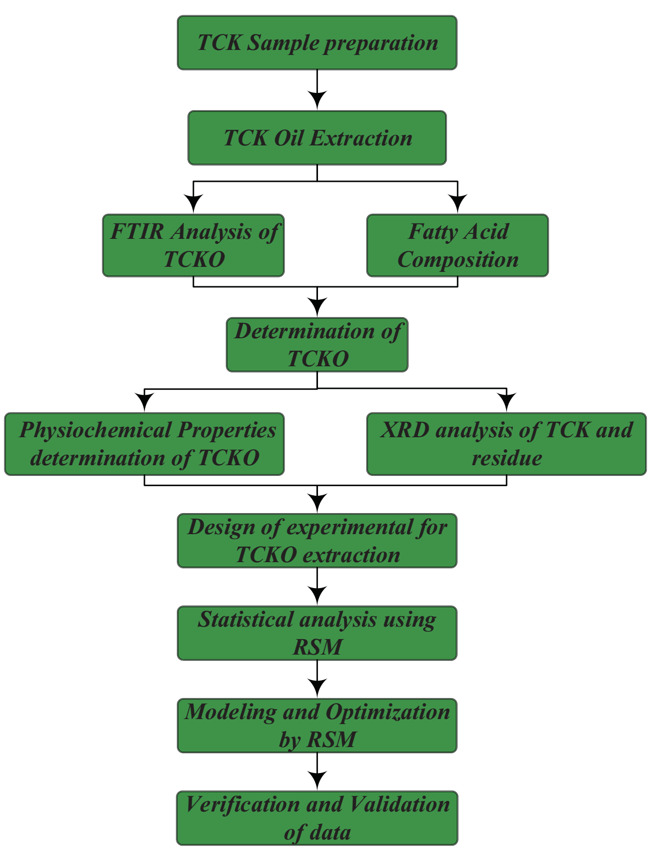

Fig. 1. Flow chart of the processes/steps used in this study.

3. Results and discussion

Oil was extracted from TCK using solvent extraction method. The efficiency of RSM and ANN were evaluated in the optimization and modeling studies.

3.1. TCK oil extraction process: modeling and parameters optimization using RSM

Table 2 shows the CCD experimental design as well as, the experimental and predicted TCK oil yields values. Using Minitab 17.0 software, the full regression model equations' terms and statistical significance were determined. The second-order polynomial regression model that best described the process as a function of actual values of temperature (X_1), particle size (X_2) and extraction time (X_3) for TCK oil extraction is given by Eq. (5).

$$\begin{aligned} TCK\%Yield = {} & 39.97+4.75X_1-11.67X_2+17.32X_3 \\ & +3.55X_1^2-0.47X_2^2-6.02X_3^2-0.74X_1X_2 \\ & +0.78X_1X_3-7.52X_2X_3 \end{aligned} \tag{5}$$

Similarly, Eq. (6) shows the model equation for TCK extraction, after all insignificant terms have been removed. These insiginificant terms were removed based on their *p*-values in the ANOVA results in Table 3. Table 3 shows the analysis of variance table for TCK oil extraction process. These removed insignificant terms are those terms whose p-values were >0.05.

$$\begin{aligned} TCK\%Yield = {} & 39.97-4.75X_1-11.67X_2 \\ & +17.32X_3-6.02X_3^2-7.52X_2X_3 \end{aligned} \tag{6}$$

ANOVA test was used to evaluate the statistical significance of the model equation and the model terms (Table 3). A calculated F value, > the F-table (Critical F value) value implies that the model was adequately fitted to the experimental data (Talib et al., 2016; Wu et al., 2016; Bezerra et al., 2008; Bai et al., 2015). Based on a 95% confidence

Table 3
Analysis of variance (ANOVA) for response surface quadratic model of TCKO extraction.

Source of variable	SS	Df	Coeff. (β)	SE coeff.	MS	F-value	P-value	T-value
Model (C.)	2455.52	9	39.97	1.13	272.84	35.14	0.000	35.27
X_1	112.8	1	4.75	1.25	112.8	14.53	0.003	3.81
X_2	680.93	1	−11.67	1.25	680.93	87.71	0.000	−9.37
X_3	1499.89	1	17.32	1.25	1499.89	193.2	0.000	13.90
X_1^2	29.73	1	3.55	2.04	23.4	3.01	0.113	1.74
X_2^2	0.00	1	−0.47	2.04	0.4	0.05	0.825	−0.23
X_3^2	67.19	1	−6.02	2.04	67.19	8.66	0.015	−2.94
X_1X_2	0.62	1	−0.74	2.63	0.62	0.08	0.783	−0.28
X_1X_3	0.68	1	0.78	2.63	0.68	0.09	0.774	0.30
X_2X_3	63.68	1	−7.52	2.63	63.68	8.2	0.017	−2.86

[C] Constant.
[SS] Sum of squares.
[Df] Degree of freedom.
[Coeff.] Coefficients.
[SE Coeff.] SE Coefficient.
[MS] Mean square.
X_1: temperature; X_2: particle size; X_3: time.
p < .01 highly significant; 0.01 < p < .05 significant; *p* > .05 not significant.
Model Tabulated $F_{0.05,9,9}$ value is 3.19. Hence, F > 3.19 significant; F < 3.19 insignificant.
Model terms Tabulated $F_{0.05,1,9}$ value is 5.12. Hence, F > 5.12 significant; F < 5.12 insignificant.

level, the model for TCK was found to be adequate as its calculated F value (35.14) was > the table $F_{0.05,9,9}$ value (3.19) (see Table 3). Moreover, the terms in the model were also tested for significance at 95% confidence level. The tabulated $F_{0.05,1,9}$ value was 5.12. Therefore, the terms in the model, X_1, X_2, X_3, X_3^2, X_2X_3 were all significant for TCK models.

The p-value provide details as to whether a statistical hypothesis is significant or not and how significant it is. When the calculated p-value is <0.05, based on 95% confidence level, the evidence against null hypothesis H_0 is stronger (Talib et al., 2016). Therefore, the model for TCK oil extraction was found to be significant as its p-value was = 0.000 and < 0.05. Similarly, the terms (X_1, X_2, X_3, X_3^2, X_2X_3) in the TCK model were all significant ($p < .05$) (Table 3).

The characteristics parameters that may express the quality (goodness of fit) of the proposed second order polynomial model (Eq. (6)), are the coefficient of determination (R^2) and adjusted R^2 (Adj-R^2) (Mazaheri et al., 2017). From Table 4, the value of R^2 for TCK oil extraction process is 96.94%. The closer the R^2 to 1 (100%), the better the fit. Similarly, the value of the R^2 Adj. is 94.18%. The value of R^2, shows significant closeness to 1, hence, indicate good fit. The values of the predicted model R^2 and the adjusted R^2 were both found to be very high (Table 4). The closeness between R^2 and Adj-R^2 values, in addition to the lower value of Adj-R^2 compared to the R^2 value, for the model, indicates goodness of data fit (Mazaheri et al., 2017; Wu et al., 2016; Mirhosseini and Tabatabaee, 2012; Samaram et al., 2015; Betiku and Ajala, 2014; Li et al., 2009; Agu et al., 2018; Agu et al., 2015). In other words, the better the empirical model fit the experimental data (Danbaba et al., 2015).

Since the model (Eq. (6)) was statistically significant, it was used to predict the optimum condition values for obtaining maximum oil yield of TCK sample using Minitab 17.0 software. The optimum conditions for maximum TCK oil yield with respect to the proposed model equation were 55 °C, 0.5 mm particle size and 150 min. At these conditions, the RSM theoretically obtained oil yield was 62.92%, which was validated as 60.34%. The validation experiment was done by carrying out three independent experimental replicates and the average recorded. The closeness of the validated and predicted TCK oil yields, confirmed the adequacy and validity of the models.

3.2. TCK oil extraction process: modeling and parameters optimization using ANN

The artificial neural network architecture was developed using Neural Network Toolbox of MATLAB R2017a software package. In this study, several neural network architecture and topologies for the estimation and prediction of TCK oil yield were tested. Thereafter, the best ANN model was chosen. Learning algorithm and transfer function effects were studied through the successful training of neural network model. This was done by employing the different learning algorithms and transfer functions of ANN. The neural network learning rate is affected by the transfer function which also aids its performance (Betiku and Ajala, 2014). A series of topologies were examined so as to determine the optimum number of neurons in the hidden layer and these values were varied from 10 to 15. Also, R^2 and RMS were used to measure the predictability of the network. Hereafter, the best topology that is 3 – 12 – 1, which has three inputs (temperature, particle size and extraction time), twelve neurons as the optimum, and one output (% oil yield), was chosen and used to predict the oil yield of TCK sample. This was chosen due to the fact that the R^2 values of both training and testing sets, for the studied sample, were very close or approximately 1.0; while the RMS error values were the least of all the topologies. The architecture of the chosen learning algorithm is depicted in Fig. 2. The Multilayer Full FeedForward (MFFF) Incremental Back Propagation network, coupled with sigmoid function consisting of 3 – 12 – 1 topology, was the ANN model used in this study. 10,000 iterations were used for the learning. Fig. 3(a) shows a comparison of the ANN predicted and experimental values. From the figure, it could be seen that the ANN was able to predict known data response, that is, the data used for training. ANN was also able to predict the unknown data, that is, the data that was not used for training. In other words, the ANN model can be used efficiently for the description of the relationship between input variables (temperature, extraction time and particle size) for TCK oil extraction. The optimum conditions established for maximum TCK oil yields using the network model were 55 °C, 0.5 mm and 150 min. At these conditions, the theoretical oil yield was 60.39%. This value was validated as 60.40%. The validation experiments were done by carrying out three independent experimental replicates and the average value noted. The closeness of the validated and predicted oil yield confirmed the adequacy and validity of the ANN model.

3.3. Performance comparison of ANN and RSM

In order to check the models obtained from RSM and ANN, both R^2 and RMS were evaluated in the studied TCK sample (Table 5). The experimental and the predicated (RSM and ANN) models oil yields, the models residuals and RMS values for TCK oil extraction, are presented in Tables 5. The obtained results indicated that both optimization tools gave good predictions as supported by high values of the R^2 and small values of RMS. The RSM's R^2 value was 96.94% (0.9694), while the ANN's R^2 value was 0.9996 [see Fig. 3(a)]. However, ANN showed observable superiority over RSM due to its higher R^2 and smaller RMS obtained values. Also, the residual values obtained by calculating the difference between the experimental and the models' (RSM and ANN) values, for the sample studied, indicated that the ANN had preeminence over RSM, with smaller obtained residual values. Furthermore, data

Table 4
Analysis of variance of regression for response surface model of TCKO extraction.

Regression	R^2 (%)	Adj. R^2 (%)	F-value	P-value
Linear	90.54		98.48	0.000
Quadratic	3.83		4.16	0.037
2-way interaction	2.57		2.79	0.095
Total model	96.94	94.18	35.14	0.000

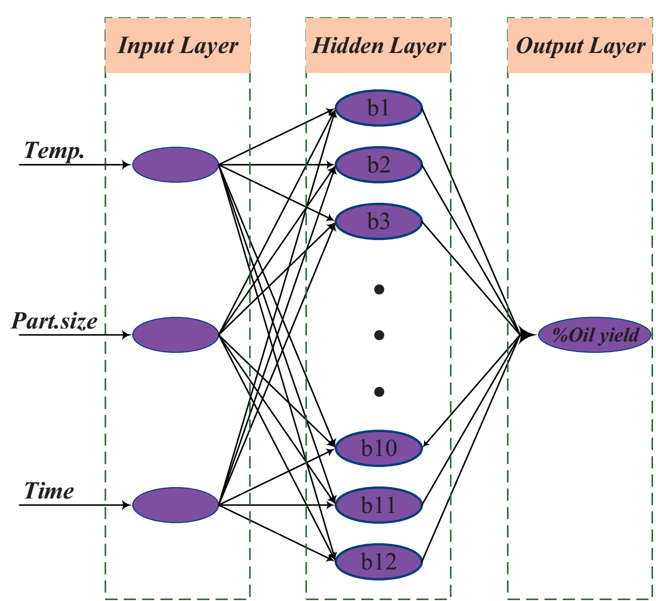

Fig. 2. Artificial neural network (ANN) model for the oil extraction from the *Terminalia catappa* L. kernel (TCK) sample.

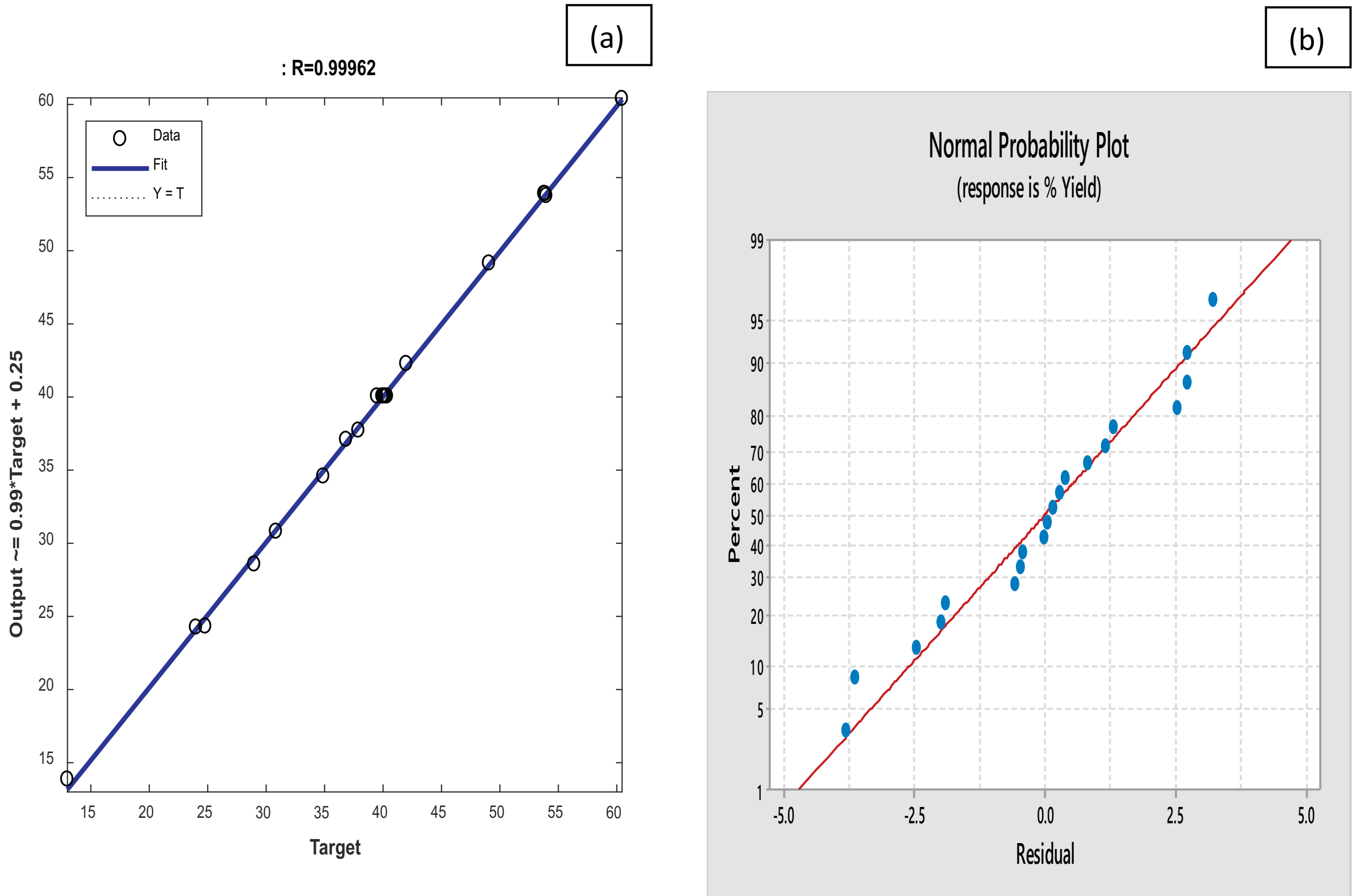

Fig. 3. Graph of Exp. value against (a) ANN predicted value and (b) RSM predicted value for TCK oil extraction.

fitting of the models were evaluated in the entire studied samples, with ANN [Fig. 3 (a)] demonstrating better fitting than RSM [Fig. 3 (b)]. In conclusion, ANN was better than RSM in the modeling and optimization studies of TCK oil extraction.

3.4. Physicochemical characterization of TCKO

The physicochemical characteristics of TCKO are shown in Table 6. The oil yield of TCK was found to be 60.45% (in mass). This value was

Table 5
Experimental and predicted RSM and ANN TCK oil yields with their residuals and RMS error values.

Run	Temp. (°C) X_1	Part. Size (mm) X_2	Time (min) X_3	Oil Yields (%)			Residual		RSM & ANN comparison by RMS error	
				Exp.	RSM	ANN	RSM	ANN	RSM	ANN
1	−1.000	1.000	−1.000	24.80	21.60	24.32	3.20	0.48	0.0912	0.0137
2	1.000	−1.000	−1.000	34.90	35.48	34.58	−0.58	0.32	0.0118	0.0065
3	0.000	0.000	0.000	39.50	39.97	40.05	−0.47	−0.55	0.0084	0.0098
4	1.000	1.000	1.000	42.00	42.43	42.27	−0.43	−0.27	0.0072	0.0045
5	−1.000	−1.000	1.000	53.98	55.96	53.75	−1.98	0.23	0.0259	0.0030
6	1.000	−1.000	1.000	60.45	62.92	60.39	−2.47	0.06	0.0289	0.0007
7	0.000	0.000	0.000	40.00	39.97	40.05	0.03	−0.05	0.0005	0.0009
8	0.000	0.000	0.000	39.95	39.97	40.05	−0.02	−0.10	0.0004	0.0018
9	1.000	1.000	−1.000	28.99	26.27	28.56	2.72	0.43	0.0663	0.0105
10	−1.000	1.000	1.000	37.90	36.58	37.71	1.32	0.19	0.0246	0.0035
11	−1.000	−1.000	−1.000	30.85	29.69	30.80	1.16	0.05	0.0266	0.0011
12	0.000	0.000	0.000	40.11	39.97	40.05	0.14	0.06	0.0025	0.0011
13	−1.633	0.000	0.000	36.85	38.77	37.08	−1.92	−0.23	0.0368	0.0044
14	0.000	−1.633	0.000	53.89	51.17	53.88	2.72	0.01	0.0357	0.0001
15	0.000	1.633	0.000	24.01	27.83	24.25	−3.82	−0.24	0.1125	0.0071
16	0.000	0.000	1.633	53.80	51.27	53.90	2.53	−0.10	0.0333	0.0013
17	0.000	0.000	0.000	40.35	39.97	40.05	0.38	0.30	0.0067	0.0053
18	1.633	0.000	0.000	49.08	48.27	49.14	0.81	−0.06	0.0117	0.0009
19	0.000	0.000	0.000	40.25	39.97	40.05	0.28	0.20	0.0049	0.0035
20	0.000	0.000	−1.633	13.00	16.63	13.86	−3.63	−0.86	0.1974	0.0468

higher than the values reported for cotton seed (Khan et al., 2010) and soybean (Lawson et al., 2010). Hence, this is an indication of its potential economic benefit as well as possible industrial application. Similarly, oil yields of 50 and 49%, were reported for TCK obtained in Brazil by Iha et al. (2014) and Dos Santos et al. (2008), respectively. These values were less than that obtained in this work. These differences in the obtained oil yields for TCK could be attributed to factors like, geographical location, seed variety, harvest period, soil texture and method of oil extraction used (Menkiti et al., 2015; Berti et al., 2011). Consequently, TCKO could constitute an alternative source of oil for industrial application as transformer fluid because of its relatively high yield.

Table 6 also shows some important physicochemical properties of TCKO. From Table 6, it could be observed that the viscosity and acidity of TCKO were 20.29 $mm^2 S^{-1}$ and 4.73 mg KOH/g, respectively. Though, these values differ from that reported by Iha et al. (2014), with the viscosity and acidity in this work being higher and lower, respectively, than 5.5 $mm^2 S^{-1}$ and 150.9 mg KOH/g oil reported by Iha et al. (2014). In work of Dos Santos et al. (2008), they reported higher viscosity of TCKO (39.8 $mm^2 S^{-1}$). This high viscosity and the lower acid value of TCKO in this report, compared to those reported by these other authors, could be due to the breed of *Terminalia catappa* L. (Ejikeme et al., 2010). Therefore, it could be affirmed that breeds of *Terminalia catappa* L. enhance its yield and oil properties. In this work, the iodine value (IV) of TCKO (101.86 g/I_2/100g oil) was higher than the 83.92 g/I_2/100g oil and 70.41 g/I_2/100g oil reported by Dos Santos et al. (2008) and Monnet et al. (2012), respectively; but lower than 135.50 g/I_2/100g oil reported by Adepoju et al. (2014). The high iodine values of the oil are indication of the high level of unsaturation in the oils. Also, the density and moisture content of TCKO are 890 g/cm^3 and 2.1 mg/kg, respectively (Table 6). This density was lower than 871 g/cm^3 reported by Iha et al. (2014), while the moisture content was lower than 2.45 reported by Adepoju et al. (2014). Furthermore, the pour and flash points of TCKO are 3 and 260 °C, respectively (Table 6). This pour point value was lower than 11.5 °C reported by Orhevba et al. (2016) for *Terminalia catappa*. Similarly, the flash point in this work was higher than 208 °C, also reported by Orhevba et al. (2016). Finally, the dielectric strength (DS) value of *Terminalia catappa* kernel oil (TCKO) was 30.61 KV (Table 6). The dielectric strength is defined as the maximum electric field, which a pure material/substance can withstand under ideal conditions, without experiencing failure of its insulating properties (Derick et al., 2014). This value was lower than that of soybean oil (39 KV), but higher than that of palm kernel oil (25 KV) (Usman et al., 2012). However, the DS value of TCKO was improved with further purification and transesterification to obtain modified TCKO transformer fluid (Menkiti et al., 2017; Agu et al., 2019). This is because in generally, oils with high DS is required for proper insulation and cooling in the transformer, since its insulating properties are not easily lost with time during transformer usage (Menkiti et al., 2017; Agu et al., 2019).

Table 6
Physicochemical properties of TCKO.

Oil property	TCKO	Standard spec. For mineral TO	Standard Methods
Oil yield (%)	60.45	–	AOAC 920.85
Dielectric strength (KV)	30.61	40–60	IEC 60156
Viscosity ($mm^2 s^{-1}$)	20.29	10	ASTM D445
Acidity (mg KOH/g oil)	4.73	0.5	AOAC 969.17
Density at 20 °C (gm^{-3})	890	870	AOAC 985.19
Iodine Value (g/I_2/100g oil)	101.86	–	AOAC 993.20
Moisture Content (mg/kg)	2.1	<20	ASTM E203
Flash Point (°C)	260	152	ASTM D93
Pour Point (°C)	3	−48	ASTM D97

Table 7
Fatty acid composition of TCKO.

Fatty acid component	Fatty acid type	% concentration
C14:0 (Myristic acid)	Saturated	7.04
C16:0 (Palmitic acid)	Saturated	9.62
C16:2 (Hexadecadienoic acid)	Unsaturated	1.00
C17:0 (Margaric acid)	Saturated	0.81
C18:1 (Oleic acid)	Unsaturated	81.40
C20 (Arachidic acid)	Saturated	0.13
Saturated fatty acids (%)		17.60
Unsaturated fatty acid (%)		82.40
Total (%)		100

3.5. Fatty acid composition of TCKO

The fatty acid composition of *Terminalia catappa* kernel oil (TCKO) is presented in Table 7, while its fatty acid profile is presented in Fig. 4. It is important to note that one of the most important parameter that affects the fatty acid, as well as oil properties is the degree of unsaturation. The iodine value gives an indication of this degree of unsaturation of oils (Ekpa and Isaac, 2013). Therefore, the iodine value of TCKO sample (see Table 6), indicated that it is semi-drying oil. This was attributed to its iodine value that falls within the range of 90 to 130 g/I_2/100g oil, required for semi-drying oils (Ekpa and Isaac, 2013; Guner et al., 2006). From the results in Table 7, TCKO had over 80% of its composition as unsaturated fatty acid, with only about 17.60% as the saturated fatty acid. The saturated and unsaturated fatty acid compositions of TCKO were 17.60% and 81.40%, respectively. This is an indication that the oil sample was highly unsaturated. Hence, the implication of this is that the oil sample cannot be used in its present state as an industrial fluid, especially as transformer oil. This is attributed to their high level of unsaturation. From the fatty acid profile, it was found that TCKO is composed of six (6) fatty acids. These fatty acids are myristic, palmitic, hexadecadienoic, marganic, oliec and arachidic acids. Oleic acid was the predominant unsaturated fatty acid with 81.40% of the total unsaturated fatty acid composition. This high level of unsaturated fatty acid in the TCKO is an indication of its high oxidation tendency (Menkiti et al., 2015). There is therefore need to improve on its percentage saturated fatty acid composition using chemical modification methods. This is to ensure that more stable oil is obtained for industrial purposes (Menkiti et al., 2017; Agu et al., 2019). On the other hand, myristic, palmitic, marganic and arachidic acids are the saturated fatty acid present in TCKO. From available information in literature, TCKO is highly composed of unsaturated fatty acids, with Oleic acid always present in high concentration. For instance, Iha et al. (2014), Dos Santos et al. (2008), Janporn et al. (2014), Adepoju et al. (2014) and Monnet et al. (2012), reported the that saturated and unsaturated fatty acid composition of *Terminalia catappa* kernel oil, were (34% and 64.5%), (40% and 60%), (40.08% and 54.75%), (39.82% and 60.18%) and (40.22% and 60.14), respectively. The difference in the fatty acid composition of TCKO in this work, and those of the previous works, could be attributed to factors, like geographical location and variety (Menkiti et al., 2017; Ejikeme et al., 2010). Therefore, there is the need for the modification of the TCKO, in other to improve on its saturation level, prior to its possible industrial application as transformer fluid (Menkiti et al., 2017; Agu et al., 2019).

3.6. FTIR analysis of Terminalia catappa L. kernel oil (TCKO)

Fig. 5 shows the FTIR spectrum of TCKO sample. The result in Fig. 5 was analyzed and compared with known signature of identified materials in the FTIR library (Barbara, 2004). For the TCKO sample, Fig. 5, the peak center at 810.7829 cm^{-1}, is characteristics of NH_2 wagging and twisting, indicating the presence of amines. The peak at 1207.488 cm^{-1} is a characteristic of C – O stretching, indicating the

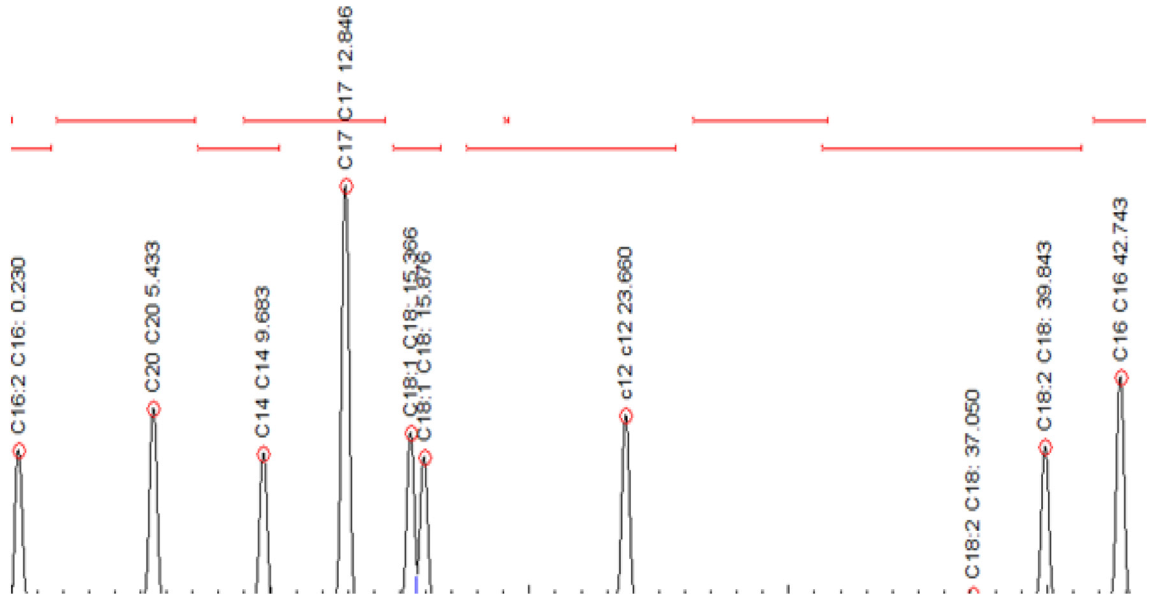

Fig. 4. Fatty acid composition profile of *Terminalia catappa* L. kernel (TCKO) sample.

presence of alcohol, phenol and anhydrides, which are oxygen-containing compounds. For the peak at 1914.455 cm^{-1}, it is a characteristic of overtone and combination bands, indicating the presence of aromatic compounds. The peaks centers at 1454.543 $cm^{-1}$and 2083.745 cm^{-1}, are characteristics of first overtone N – H stretching/ first overtone O – H stretching and combination N – H stretching/

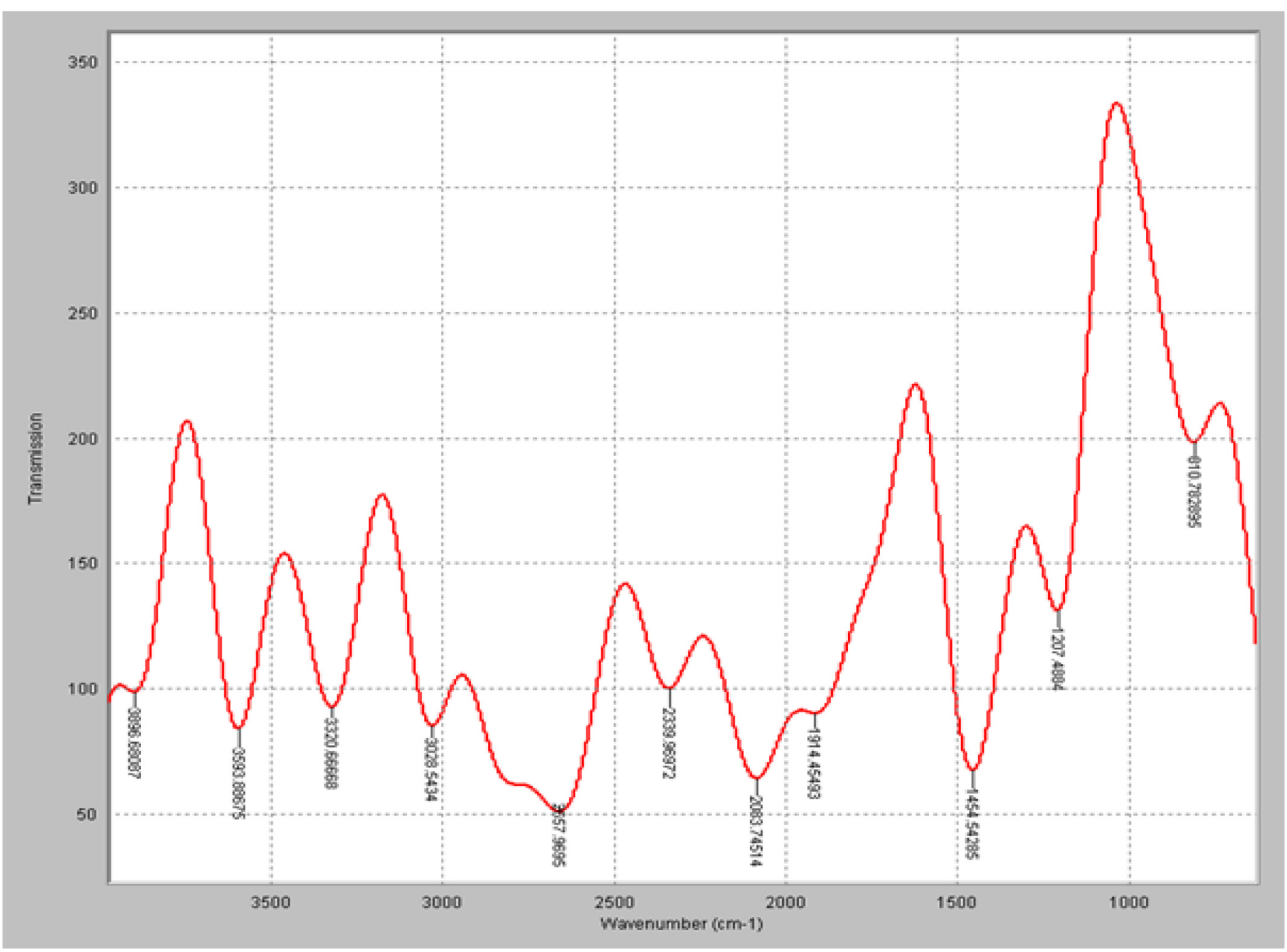

Fig. 5. Fourier Transform Infrared (FTIR) spectrum of *Terminalia catappa* L. kernel (TCKO) sample.

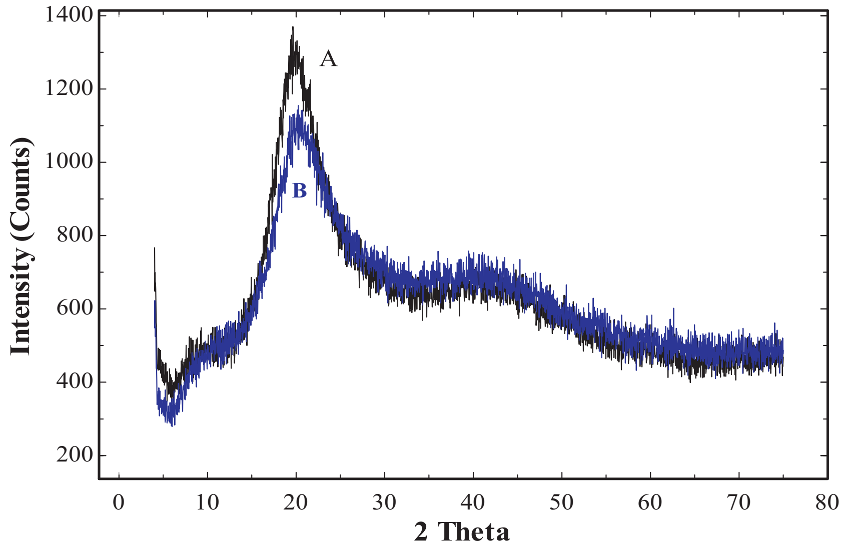

Fig. 6. XRD Pattern of milled TCK. A: before, and B: after extraction.

combination O – H stretching, respectively; which are indications of the presence of organic compounds. In like manner, the peak at 2339.97 cm^{-1} is characteristics of phosphorous acid and ester P – H stretching, indicating the presence of phosphorus compounds. The peak ranges at 2657.969 cm^{-1} and 3028.543 cm^{-1} are characteristics of O – H stretching (carboxylic acids), which indicates the presence of oxygen-containing compounds. Furthermore, the peaks at 3320.667 cm^{-1} and 3593.887 cm^{-1} are characteristic of alcohol O – H stretching, indicating the presence of alcohols, which are oxygen-containing compounds. Finally, the peak at 3896.681 cm^{-1} is far beyond the infrared band of 3700 wavenumber (cm^{-1}), for organic compounds and could not be identified.

3.7. X-ray diffraction (XRD) spectra of Terminalia catappa L. kernel (TCK) and its residue

X-ray diffractometry is widely used to reveal the characteristics of the crystalline structure of substances, such as, starch granules, milled seeds and nuts samples (Kaptso et al., 2014). The X-ray diffraction spectra from milled *Terminalia catappa* L. kernel (TCK) sample obtained both before and after extraction are presented in Fig. 6. The XRD patterns suggest the crystalline nature of the samples with diffracting peaks at different degrees and intensity. For the TCK sample, the XRD spectra of both milled and residue after extraction, indicated prominent peaks appearing at almost the same 2θ angle (positions), but at different intensities. This indicates slight difference in the degrees of the crystallinity of the samples, both before and after extraction. Similar observations were noticed by Adama et al. (2014) and Gonçalves et al. (2014), in the crystalline nature of different starches obtained from different sources. From Fig. 6, spectra A and B represent the XRD pattern of milled TCK, before and after extraction, respectively. In the XRD pattern, the TCK samples before (spectrum A) and after (spectrum B) extractions, both have two major characteristics peaks. For the spectrum A, the peaks at $2\theta = 19.66°$ and $40.57°$, have intensities of 1353 and 717, respectively. On the other hand, spectrum B has peaks at $2\theta = 21.00°$ and $41.01°$, with corresponding intensities of 1110 and 754, respectively. The difference in both the peaks and the corresponding intensities for the TCK samples, before (spectrum A) and after (spectrum B) extraction, could be attributed to the damage caused by the solvent effect on the TCK sample structure (spectrum A). Thus, destroys the order (crystalline areas) in the TCK sample (spectrum A) structure, giving rise to spectrum B (after extraction), hence, the change in crystallinity (Agu, 2019).

4. Conclusions

It could be concluded from this study that the RSM and ANN were efficient tool for the modeling and optimization of TCK oil extraction. TCK oil extraction was successfully optimized using RSM and ANN with oil yield of approximately 60.45%. However, ANN was a better and more effective optimization tool than RSM, due to its higher R^2 and lower RMS values. The physicochemical properties of TCKO indicated its potential for industrial application, especially as transformer fluid. The fatty acid composition indicated that TCKO was highly unsaturated. Furthermore, the presence of O – H, N – H, and C – O functional groups, in the FTIR results, are indications of oxygen containing compounds, hence, would enhance TCKO biodegradability. Finally, the XRD results of TCK samples obtained both before and after extraction, showed difference in their peaks and corresponding intensities, due to the damage effect of solvent.

CRediT authorship contribution statement

Chinedu Matthew Agu Writing - review & editing. **Matthew Chukwudi Menkiti** Writing - review & editing. **Ekwe Bassey Ekwe** Writing - review & editing. **Albert Chibuzor Agulanna** Writing - review & editing.

Abbreviations

ANN	artificial neural network
ANNs	artificial neural networks
ANOVA	analysis of variance
AOAC	Association of Official Analytical Chemists
CCD	central composite design
DS	dielectric strength
FTIR	Fourier Transform Infrared
GC	gas chromatography
IBP	Incremental Back Propagation
MFFF	Multilayer Full Feedforward
MNFF	Multilayer Normal Feedforward
RSM	root mean square
RSM	response surface methodology
TCK	*Terminalia catappa* kernel
TCKO	*Terminalia catappa* kernel oil
XRD	X-ray diffraction

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