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Microwave mediated production of FAME from waste cooking oil: Modelling and optimization of process parameters by RSM and ANN approach

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ABSTRACT

Fatty acid methyl esters (FAME) were synthesized from the waste cooking oil (WCO) by direct transesterification with methanol. The WCO was pretreated to increase the efficiency of FAME production with 1% of potassium methoxide as a catalyst. The free fatty acid of raw source was reduced using 2% of activated charcoal as adsorbent. The process variables were optimized using Box-Behnken design of Response Surface Methodology (RSM), and developed Artificial Neural Network (ANN) model to predict the FAME yield. The highest percentage of conversion (95%) was achieved at optimum conditions, for the catalyst concentration of 1%, alcohol to oil ratio of 6:1, temperature of 75 °C and time of 60 s. The yield of biodiesel was estimated by higher R^2 values of RSM (0.98) and ANN (0.99), respectively. The produced FAME from pretreated WCO at 75 °C by microwave irradiation was examined by proton nuclear magnetic resonance (^{1}H NMR), carbon nuclear magnetic resonance (^{13}C NMR), Fourier transform infrared (FT-IR) spectroscopy, and gas chromatography-mass spectrometry (GC–MS). The esters exhibited their two characteristically strong absorption bands arising from carbonyl (C═O) at 1741 cm^{-1} of fresh oil, and C─O stretching between 1300 and 1000 cm^{-1} in the FT-IR spectra. A representative spectrum of ^{13}CNMR for the FAME from WCO was confirmed the presence of methyl esters in the biodiesel of ester carbonyl (─COO─) and C─O at 174.2 ppm and 51.4 ppm, respectively. The characteristic peak of ^{1}H NMR at 1.00 ppm of methoxy protons was observed as a singlet at 3.67 ppm, which proved occurrence of the methyl ester (CH_3COOR). In addition, a significant weight loss at 139 °C was observed through thermogravimetric (TG) analyses of FAME.

1. Introduction

Availability source of fossil fuel energy is getting decreased continuously due to its large consumption in the worldwide. The necessity for an alternative to fossil fuels is increasing day by day due to the irreplaceable effects on the environment in the form of air pollution. Consequently, there is a demand to develop new energy sources that should be renewable, ecofriendly, and economically feasible [1]. Biodiesel is one of the best alternatives to fossil fuel energy source [2]. Biodiesel is produced from triglycerides of oil by transesterification in the presence of catalyst base *e.g.* NaOH, KOH, acid like H_2SO_4 and enzyme [3–6]. For commercial purpose, base catalyst and immobilized enzyme catalyst have been used to produce fatty acid methyl ester (FAME) of triglycerides. This FAME is nontoxic, biodegradable and less carbon dioxide emission [7]. Biodiesel is derived from fats and oils either by chemical or biochemical or enzymatic method [8]. Various feedstock edible and non-edible oils are proven diesel substitutes [9–12]. Triglycerides can be converted into biodiesel by blending, transesterification, pyrolysis, and micro emulsions. Transesterification method is commonly used to reduce the viscosity of oil and addition of activated carbon [13–16]. The transesterification process is energy efficient and a quick technique to produce biodiesel from different feedstock [16–21].

Vegetable oil can be chosen as an alternative source for the biofuel production. Biodiesel is an impressive and essential one that can be produced through the vegetable oil. The waste cooking oil (WCO) can be used for the production of biodiesel owing to the need of food security, cost effective alternative, and above all the release of greenhouse gases. Currently, the generation of WCO from restaurant, hotels, household and industry is increasing and become the global issue in and

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around the world. The WCO is discharged into the drain, which integrate with food chain of environment and cause several problems to the living species [22,23]. WCO is the best interest for biodiesel production and, it is approximately three times cheaper than fresh oils [24]. A transesterification reaction by conventional heating such as convection, conduction and radiation of heat is energetically inefficient [25]. Microwave irradiation is a best strategy to accelerate transesterification reaction involved in cooking oil conversion [26]. However, WCO has various troubles including saponification reaction during conversion and causes a huge effect in production. Therefore, the WCO is pretreated, and the debris, free fatty acid (FFA), acid value, moisture content is reduced, which are the causes of reduces in efficiency of the biodiesel. The pretreatment can be carried out by the addition of adsorbent. Activated carbon and silica gel mixture can able to reduce acid value up to 53.9% [26].

In several studies, microwave heating technique is successfully applied to the biodiesel production from a variety of renewable feedstock in batch mode [27–30]. The microwave interaction with the reagents (triglycerides and methanol) to accelerate thetransesterification reaction already well-established and 95% yield obtained at very short reaction time [31]. Microwave assisted transesterification of different feedstock such as rapeseed oil, grease oil, cotton seed oil and WCO are reported elsewhere [32–34].

This present study deals with the synthesis of FAME from WCO by potassium methoxide catalyzed transesterification using microwave heating with methanol and the potential evaluation of WCO for the commercial production of biodiesel. A comparative study was made in the characteristics among fresh oil, cooked oil, pretreated oil and biodiesel and their data were interpreted in systematic manner [35–39]. Several standard properties namely, FFA content, acid value, moisture content, colour, specific gravity, iodine value, saponification value, refractive index and peroxide index were characterized, compared and correlated. The produced biodiesel was characterized by physico-chemical properties and its compositions were determined by ^{1}H NMR, ^{13}C NMR, GC–MS, FT-IR and TG analysis [39–48].

Process optimization using multi-variant techniques have employed widely through mathematical model of Response Surface Methodology (RSM) [49]. A mathematical correlation between processes variables are developed using Box-Behnken design of experiments to obtain optimal yield of biodiesel. Nevertheless, RSM, which is nonlinear process, shows limitation towards variables range [50–53]. The limitation in RSM has been fulfilled by Artificial Neural Network (ANN) with any complex nonlinear process to make appropriate model without complex equation. Hence, in this study, the microwave mediated production of FAME from WCO process parameters are optimized by RSM and ANN method.

2. Experimental

2.1. Materials and methods

WCO was collected from restaurants located in Tiruvannamalai, a vicinity of Tamil Nadu in India. Methanol (99.9% purity), potassium hydroxide (99.9% purity), activated charcoal, silica gel and phenolphthalein were procured from Sigma-Aldrich. All the chemicals used were analytical grade. A domestic microwave oven (Samsung, Model CE104VD) was used for the heating and treatment of the WCO.

2.2. Production of FAME from WCO

2.2.1. WCO pretreatment

WCO was heated at 110 °C to remove the moisture followed by filtered with Whatman filter (10 μm) paper to eliminate any presented inorganic residues and suspended matters. Filtered oil was treated with 1–5% (w/v) activated carbon for five days and stirred for 30 min at speed of 60–70 rpm. Afterwards, the sample was filtered through Whatman filter paper to remove activated carbon from the WCO.

2.2.2. Transesterification of WCO

Before starting the reaction, 1 L of pretreated WCO was heated at 75 °C using microwave oven. For the catalyst preparation, 6:1 vol ratio of methanol to oil and 1% KOH (w/v) were used. Further, the transesterification reaction was carried out using WCO with 200 mL of potassium methoxide as catalyst at 80 °C at constant stirring speed about 600 rpm for 10 min to obtain the complete conversion of oil to FAME. After the reaction, the content was transferred to separating funnel for the separation and allowed to settle two phases. From the separating funnel, the glycerin layer was removed, fatty methyl ester was collected, followed then FAME was washed with hot water to remove excess residual FFA. Finally, the sample was dried to remove excess methanol of reaction mixture. The yield was calculated using Eq.1.

$$Yield\ (\%) = \frac{Amount\ of\ biodiesel\ produced\ (g)}{Amount\ of\ total\ oil\ (g)} \times 100 \tag{1}$$

2.3. Experimental design

2.3.1. Box-Behnken design of response surface methodology (RSM)

The biodiesel synthesis using potassium methoxide as a catalyst was designed and optimized using Box-Behnken design for RSM, which is a mathematical statistical regression model. This process requires adequate information of factors, levels and responses. Experiments were carried out according to Box-Behnken design that involves 4 variables and 3 levels, which consists of 29 experimental runs. The variables and its levels for Box-Behnken design for RSM used in this study are shown in Table 1. The following second-order non-linear polynomial equation was fitted for the experimental data.

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i X_i + \sum_{i=1}^{n} \beta_{ii} X_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \beta_{ij} X_i X_j + \varepsilon \tag{2}$$

where Y is the response, that is biodiesel yield (%), n is the number of variables, β_0 is the model intercept term and β_i is the liner effect term, β_{ii} is the square effect term, β_{ij} is the interaction effect term, $X_i$and X_j is the level of the independent variables and ε is the random error. The statistical analyses of the experimental data were performed by Design Expert 10, Stat-Ease, Inc., Minneapolis, USA. The data were fitted with the equation represented by Eq. (2) for the yield in terms of actual values of independent variables.

2.3.2. Artificial Neural Network (ANN)

ANN is a mathematical simulating tool for complex process. It is composed of nodes called neuron and weights. The neurons are arranged in the form of interlinked single or multiple layers of independent variables and dependent variables. ANN is composed of structural components for input layer, hidden layer and output layer. The entire hidden layer is composed by interlinked nodes called neurons and interchanging their weights to adjacent neurons. The input layer receive signal from variables, and sent to hidden layer for processing by logsig activation function. The variables were used for input is given in the Table 2. The MATLAB (R2010) (The Mathworks, Inc.,

Table 1
Variables for Box-Behnken design in transesterification of pretreated WCO.

Variables	Levels		
	−1	0	+1
X_1: Catalyst (wt. %)	0.5	1	1.5
X_2: Alcohol: Oil ratio(v/v)	3:1	6:1	9:1
X_3: Time (s)	30	60	90
X_4: Temperature (°C)	65	75	85