



Synthesis and characterization of $\text{Sr}_3\text{Al}_2\text{O}_6$ nanocomposite as catalyst for biodiesel production



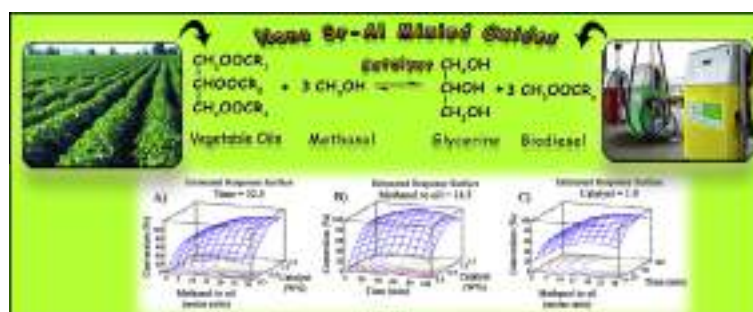
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HIGHLIGHTS

- Sr–Al mixed oxides nanoparticles prepared by sol–gel method as catalyst.
- The prepared catalyst was used for soybean oil transesterification to produce biodiesel.
- Reaction parameters were optimized by using RSM based on Box–Behnken center–united design.
- The conversion under optimized reaction conditions was $95.7 \pm 0.5\%$.

GRAPHICAL ABSTRACT



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ABSTRACT

Sr–Al mixed oxides nanocomposite as $\text{Sr}_3\text{Al}_2\text{O}_6$ was prepared by sol–gel method and used as a basic heterogeneous catalyst for transesterification of soybean oil to methyl esters (biodiesel) by methanol. The prepared catalyst was characterized using X-ray diffraction (XRD), Transmission electron microscope (TEM), thermogravimetric analysis (TGA/DTA), temperature-programmed desorption (TPD) and nitrogen adsorption–desorption techniques. The response surface methodology (RSM) based on the Box–Behnken design was employed to investigate the effects of methanol to oil molar ratio, reaction time, catalyst amount and specifically the effect of interaction between process variables on the conversion of oil to biodiesel. Results from this study revealed that individual as well as bilateral variables interactions significantly affect the yield of biodiesel. With this information, it was found that utilization of methanol to oil molar ratio of 25 and 1.3 wt.% of catalyst within reaction time of 61 min and gave the biodiesel yield of $95.7 \pm 0.5\%$.

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1. Introduction

The use of biodiesel as a replacement of fossil fuels is becoming increasingly popular nowadays due to global energy crisis. Biodiesel, a renewable, nontoxic and biodegradable is a mixture of fatty acid methyl esters (FAMES) produced by transesterification of triglycerides with methanol or other short chain alcohols in the presence of an appropriate catalyst (Dias et al., 2012; Gombotz et al., 2012; Kazemian et al., 2013). Transesterification can be

accomplished by acidic or basic catalysts. The latter is the most common, since the process is faster and the reaction conditions are moderated (Kazemian et al., 2013; Vicente et al., 2004). Traditionally, NaOH or KOH have been used as homogeneous catalysts for commercial transesterification processes. Methyl esters are produced with high yield under these basic catalyst conditions. However, these catalysts require additional neutralization and separation steps for the final reaction mixture, thus leading to a series of environmental problems related to using excessive amounts of water and energy (Gombotz et al., 2012; Meng et al., 2013). Because of these problems, a great deal of interest has recently been stimulated by the use of heterogeneous catalysts. The non-corrosive and environmentally benign heterogeneous

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catalysts are ecologically and economically important in catalysis field systems with no disposal problems. The reusability and heterogeneous catalysts lead to easier and faster product separation by filtration and better product purity since water washing step can be eliminated (Kawashima et al., 2008; Semwal et al., 2011).

So far, many efforts have been devoted toward the design of heterogeneous catalytic systems effectively applied to the transesterification of vegetable oils. Examples include impregnation alkaline metal salts on microporous material zeolites (Dang et al., 2013; Kusuma et al., 2013), mesoporous materials like MCM-41 (Kazemian et al., 2013), layered materials like clays (Rashtizadeh et al., 2010) and inorganic oxides (Chen et al., 2011; Ding et al., 2011; Evangelista et al., 2012; Li et al., 2011). Other applied catalysts include hydrotalcites (Gomes et al., 2011; Sankaranarayanan et al., 2012), alkaline earth metal oxides (Mootabadi et al., 2010) and inorganic mixed oxides (Zabeti et al., 2009; Rashtizadeh and Farzaneh, 2013). Alkaline earth metal oxides listed among inorganic oxides are biodiesel production catalysts due to their high basicities and activities. Compared to CaO and MgO, whereas SrO has demonstrated excellent catalytic activity based on the experimental results due to its strong basicity (Mootabadi et al., 2010), but the partial solubility in the biodiesel product is a challenging problem.

Based on the previous studies, many factors affect the biodiesel production process using short-chain alcohols and an alkali catalyst (Peña et al., 2013). Experiments designed by changing one factor at a time can be daunting by investigation of a larger number of experiments. Moreover, the reaction system will be poorly understood since more than one variable can simultaneously influence the system and nearly impossible to achieve the true optimal condition. As such, utilization of response surface methodology (RSM) a powerful tool for the optimization of chemical reactions and/or industrial processes (Morgan, 1991) is promising. The main advantages of this method include (1) understanding of how the process variables affect the selected process response; (2) determination of any possible interrelationship among the process variables; and (3) characterization of the combined effect that all process variables may have on the process response (Domingos et al., 2008). This method has been successfully applied in the study and optimization of biodiesel production with basic or acidic heterogeneous catalysts based on the central composite design (CCD) or Box–Behnken design (Baroutian et al., 2011; Yee et al., 2011; Yin et al., 2012). Compared to CCD, the Box–Behnken design exhibits advantages of doing lower experiments and avoiding implementation of experiments under extreme conditions (Morgan, 1991).

In this study, Sr₃Al₂O₆ nanocomposite mixed oxide containing small particle size was prepared and successfully used as a basic heterogeneous catalyst for transesterification of soybean oil. The high catalytic activity and evaluation of the experimental factors include methanol to oil molar ratio, reaction time and catalyst amount on the biodiesel conversion using RSM based on the Box–Behnken design will be discussed in this presentation.

2. Experimental

2.1. Materials

Soybean oil was obtained from Sigma. Strontium nitrate was purchased from Aldrich. Methanol, n-hexane, isopropanol, acetylacetone (acac), aluminum isopropoxide, polyethylene glycol 20,000 (PEG) and methyl heptadecanoate (>99 wt.%) as the standard of GC were purchased from Merck Chemical Company (Darmstadt, Germany) and used without further purification.

2.2. Physical measurements

Products were quantified by gas chromatography (Agilent series 6890), equipped with a 5% phenylmethylsiloxane capillary column, helium as carrier gas and flame ionization detector (FID) using methyl heptadecanoate internal standard. The soybean oil composition was calculated by “AOCS Ce 1-62” method (O'Connor et al., 1967). Products were also identified by gas chromatography–mass spectrometry (GC–MS) (complex mass selective detector Agilent 5973 network and GC Agilent 6890 network). Thermogravimetry analysis (TGA) and differential thermal analysis (DTA) were recorded using a Mettler toledo TGA/SDTA 851 instrument. The samples were initially dried and then calcined up to 1200 °C (heating rate of 10 °C/min) in an air stream. The X-ray diffractions (XRD) patterns of samples were recorded on a Philips PW1800 diffractometer using monochromatic nickel-filtered Cu K α radiation ($\lambda = 0.15405$ nm). The X-ray generator was run at 40 kV and 30 mA and the diffractograms were recorded in the 2θ range of 4°–90°. The phases were identified using the powder diffraction file (PDF) database (JCPDS, International Centre for Diffraction Data). The total basicity of the prepared catalysts was measured by temperature-programmed desorption (TPD) method using CO₂ as the probe molecule. These experiments were carried out using BELCAT-A instrument (BEL Japan, Inc.). Pretreatment of the catalyst (100 mg each) was carried out by heating up to 960 °C with a heating rate of 15 °C/min under helium flow rate (25 mL/min) in order to remove the surface adsorbed wetting 1 h followed by cooling to room temperature, the chemisorption of CO₂ was carried out by a CO₂ flow rate of 25 mL/min. The excess of CO₂ was then desorbed at the temperature of adsorption in a helium flow rate (25 mL/min) for 30 min. Thereafter, desorption of CO₂ was carried out with helium from room temperature to 1000 °C with a ramp of 10 °C/min. Transmission electron microscopy (TEM) study was performed using a Philips CM30 operated at 300 kV. BET surface area of the synthesized catalyst was carried out according to the multipoint N₂ adsorption–desorption method using Quanta chrome instrument (NOVA 2000 series). Prior to the measurements, all samples were outgassed overnight at 300 °C. The percentages of strontium and aluminum were determined by atomic absorption chemo double beam instrument. Energy dispersive spectrometer (EDS) analysis was performed by oxford instruments INCA EDS system.

2.3. Method of catalyst preparation, general procedure

The sol–gel method was used for the preparation of Sr–Al mixed oxide nanocomposite. Aluminum isopropoxide (0.01 mol in 20 ml isopropanol) was added to the mixture of 10 ml acac and 4 g PEG. After stirring and refluxing at 90 °C for 2 h, strontium nitrate (2.57 g, 0.012 mmol) was slowly added and the mixture heated to make the gel. The gel was dried at 80 °C for 24 h, followed by calcination in air at 900 °C.

2.4. Transesterification of soybean oil, general procedure

Transesterification reactions were carried out in a 50 ml flask equipped with a condenser. The desired amount of catalyst was dispersed in MeOH with magnetic stirring. The soybean oil with the average molecular weight of 881 g/mol (calculated from the saponification value (S.V. = 190 mg KOH/g, with acid value and water content 0.4 mg KOH/g and 96.7 mg/kg respectively) (Akpan et al., 2006)) was then added and the mixture refluxed for the appropriate time. The mixture was then centrifuged and the solid separated with decantation and washed with n-hexane. After joining the filtrate to the decanted liquid, the solvent was removed under reduced pressure at 60 °C. Two layers containing methyl esters, soybean oil, mono and diglycerides in the upper and glycerol in the

lower layer were separated by a decanter. The FAME content was determined by GC using the European regulated procedure EN 14103 by dissolving the ester layer (250 mg) in n-hexane (5 mL) containing methyl heptadecanoate (10 g/L of C17 ester in n-hexane) as the internal standard. The FAME content (wt.%) was calculated using Eq. (1):

$$\text{wt.}\% = \left[\frac{\sum A_i/A}{A} \right] \times (C \times V/W) \times 100 \quad (1)$$

in which $\sum A_i$ is the total peak area of soybean oil methyl esters, A is the area of methyl heptadecanoate, with the equal response factor to those of FAME, C is the concentration in mg/ml of the methyl heptadecanoate (10 mg/mL), V is the volume in ml of the methyl heptadecanoate solution and W is the weight in mg of the sample (Granados et al., 2007).

2.5. Experimental design and optimization by RSM

A three-level-three-factor Box–Behnken design was employed to investigate the operating conditions of transesterification to obtain a high conversion to biodiesel. The studied factors were methanol to oil molar ratio, reaction time and catalyst amount. A preliminary study was carried out to investigate the level of each factor. The independent factor (X), their levels, design matrix, average obtained results with two replications (conversion to biodiesel %, Y) and their estimated are shown in Table 1. Experiments were carried out in randomized order with three replications of center point in order to obtain good error estimation. This methodology allows the formulation of a second-order equation that describes the process. The conversion of biodiesel was analyzed by the multiple regressions through the least squares method to fit the following equation (Eq. (2)):

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 \quad (2)$$

where β_0 is the intercept term, β_1 , β_2 and β_3 are linear, β_{12} , β_{13} and β_{23} interactive and β_{11} , β_{22} and β_{33} are quadratic coefficients.

Designing the experiments and their evaluation based on the analysis of variance (ANOVA) were performed using the graphic software “STATGRAPHIC Centurion XV” version 15.2.00 (StatPoint, Orlean, Warrenton, VA). Finally, the optimum conditions were found based on the response surface analysis.

3. Results and discussion

3.1. Catalyst characterization

3.1.1. TG-DTA studies

The Sr–Al nanocomposite mixed oxide was prepared starting from aluminum isopropoxide and strontium nitrate in isopropanol as solvent and PEG or acac as template.

The TG and DTA curves of Sr–Al mixed oxide are shown in Supplementary, Fig. S1. As indicated in TG, constant mass achieves at above 900 °C. The curve reveals that the decomposition starts at around 210 °C with the mass loss of 37%, corresponding to the volatilization of surface adsorbed water and isopropanol. The second stage start at 310 °C and is completed at 410 °C with weight loss of 36% due to the pyrolysis of organic groups. This peak is exothermic because of concomitant oxidation reaction taking place along with decomposition. Appearance of a peak around 580–640 °C is interpreted as the initial melting of strontium nitrate followed by decomposition to nitrogen oxides with the formation of perovskite and carbonate phases (Hosseini and Eslami, 2010). The thermal events are accompanied with a 3% mass loss. The last endothermic peak observed in the temperature range of 780–900 °C with the mass loss of 7% is due to the formation of a stable phase. For this, the catalyst was calcined at 900 °C.

3.1.2. XRD studies

The major product is $\text{Sr}_3\text{Al}_2\text{O}_6$, since XRD pattern of Sr–Al nanocomposite mixed oxide indicated in Supplementary, Fig. S2 is matched with the data reported in JCPDS file No. 24-1187. The XRD pattern of the major product shows the d values appearing at 5.600, 4.235, 2.799, 2.286, 2.118, 1.981 and 1.617, corresponding to 220, 321, 440, 444, 642, 800 and 844 reflections, respectively. The presence of the minor phase consistent with JCPDS PDF No. 5-0418 reveals the formation of SrCO_3 as the minor product.

3.1.3. CO_2 -TPD studies

To investigate the basic strength of $\text{Sr}_3\text{Al}_2\text{O}_6$ as catalyst for transesterification reaction, the catalyst density and basic site distributions were determined by temperature-programmed desorption (TPD) of CO_2 . Revealing of two desorption peaks at 388 °C and 747 °C in catalyst CO_2 -TPD spectrum profile given in Supplementary Fig. S3, is consistent with the presence of two basic

Table 1
Factor domain, design matrix and response in the Box–Behnken design for optimization of biodiesel production.

Independent factor	Key	Unites	Levels		
			Low (–)	Middle (0)	High (+)
Methanol to oil	X_1	mol/mol	3	16.5	30
Time	X_2	min	15	52.5	90
Catalyst	X_3	%	0.5	1	1.5
Run	X_1	X_2	X_3	Average obtained conversion to biodiesel (%), $n = 2$	Predicted conversion to biodiesel (%)
1	+	0	–	35.0	30.50
2	0	0	0	91.5	92.17
3	–	0	+	48.0	52.50
4	–	0	–	24.5	11.75
5	–	–	0	39.0	44.50
6	0	0	0	92.5	92.17
7	+	0	+	87.0	99.75
8	0	–	+	89.5	79.50
9	0	+	+	92.0	84.75
10	0	–	–	12.0	19.25
11	+	+	0	93.5	88.00
12	+	–	0	75.0	72.25
13	0	0	0	92.5	92.17
14	0	+	–	25.0	35.00
15	–	+	0	47.0	49.75

sites. Whereas the desorption peak displaying at 388 °C is attributed to the interaction of CO₂ with the medium basic site generated from the Sr²⁺–O^{2–} ion pairs, the peak appeared at $T_{\max} = 747$ °C is the strong basic site corresponding to the isolated O^{2–} anions located on a particular position on the mixed oxide catalyst surface (Lee et al., 2013). Accordingly, liberation of respectively 1.106 and 0.509 mmol/g and totally 1615 μmol/g CO₂ from basic sites indicates that Sr₃Al₂O₆ is a highly basic nanocomposite (Fig. S3).

3.1.4. TEM studies

The TEM micrograph exhibited in Supplementary Fig. S4 indicate that catalyst has particles sizes within 15–25 nm with the surface area of 4.355 m²/g by BET analysis. It was also found that after activation of the used catalyst by washing with methanol and calcination at 700 °C, the surface area was very closed to the fresh catalyst (4.25 m²/g).

3.2. Optimization of reaction conditions by RSM

3.2.1. Development of regression model equation

The relationship between response (conversion to biodiesel) and three reaction variables (methanol to oil molar ratio, reaction time and catalyst amount) were evaluated using RSM. Results for each point obtained based on the Box–Behnken design (Table 1). Based on the multiple regression analysis, it was found that the responses indicated in Table 1 are correlated with three independent variables using polynomial equation (Eq. (2)). The full quadratic estimative response model equation in terms of coded factors is also shown in Eq. (3):

$$Y = -128.765 + 3.02167X_1 + 1.036X_2 + 255.1X_3 - 0.0947645X_1^2 + 0.00518519X_1X_2 + 1.05556X_1X_3 - 0.00801481X_2^2 - 0.14X_2X_3 - 105.083X_3^2 \quad (3)$$

Here, Y is the conversion to biodiesel and X₁, X₂ and X₃ are methanol to oil molar ratio, reaction time and catalyst amount, respectively.

ANOVA results of the quadratic models are presented in Table 2. Whereas the data related to the statistical significance of the individual parameters of the fitted model represents a linear and quadratic terms between catalyst amount (X₃ and X₃²) and methanol to oil molar ratio (X₁ and X₁²), the quadratic term of reaction time (X₂²) has the largest effect on the conversion to biodiesel due to the high F-value and its corresponding low P-values, respectively. Besides, the linear term of extraction time (X₂) with the interaction between methanol to oil molar ratio and catalyst amount (X₁X₃) significantly affect the conversion to biodiesel.

The fit goodness of the model was investigated using correlation coefficient (R²) and residuals tests. The R² statistic indicates that the model as fitted explains 94.0419% of the variability in

conversion to biodiesel. The adjusted R² statistic, a more suitable factor for comparison of the models with different numbers of independent variables, is 91.3608%. The Durbin–Watson (DW) statistic also test the residuals to determine if there is any significant correlation based on the order in which they occur in our data file. Since the P-value 0.9153 is greater than 0.05, there is no indication of serial autocorrelation in the residuals at the 5.0% significance level. These results indicated that the proposed empirical model is significant and correctly explain the behavior of the response in the experimental domain.

3.2.2. Response surface plots

The empirical model is plotted as a three-dimensional surface representing the response (conversion to biodiesel) as a function of two factors within the experimental range considered in the center level of the other factor. The response surfaces and contour plots are shown in Fig. 1. As seen in Fig. 1A, the increasing rate of conversion % value is higher when methanol to oil molar ratio is in its high level. Similarly, at higher catalyst amount (1.5 wt.%), the yield of biodiesel increases from 48 to 87 wt.% by increasing methanol to oil molar ratio from 3 to 30. This increase could be attributed to an increase in the number of available catalytically active sites and the excess amount of methanol used concomitantly shifts the reversible transesterification reaction towards the biodiesel product. The great importance of catalyst amount in the conversion to ethyl ester was also emphasized by Yuan et al. (2008).

As indicated in Fig. 1B, the significant observed increase in biodiesel yield from 12 to 89.5 wt.% by increasing catalyst loading from 0.5 to 1.5 wt.% within shorter reaction time (15 min) is obvious. However, the increase was less pronounced at longer reaction time (90 min). In another words, increasing reaction time reduces the increasing effect of catalyst amount. This may be the result of negative interaction which was correctly reflected by the negative value of regression coefficient for X₂ and X₃ interaction in Eq. (2). The positive interaction of methanol to oil molar ratio with reaction time is also observed in Fig. 1C.

3.2.3. Optimization of biodiesel yield

The fitted model presented in Eq. (2) offers a good estimate of the experimental conditions so that the given model was employed to predict the best possible transesterification process variables in order to obtain the maximum conversion to biodiesel yield. The optimal values were found to be 25, 61 min and 1.3% for methanol to oil molar ratio, reaction time and catalyst amount, respectively. To verify the prediction of the model, the optimal reaction conditions were applied to three independent replicates for biodiesel production using Sr–Al mixed oxide nanocomposite catalyst. Obtaining 95.7 ± 0.5% conversion to biodiesel nicely close to the predicted value with less than 4% error indicates that the predicted optimum process conditions used in this study are valid.

Table 2
ANOVA for the experimental results of the Box–Behnken design.

Source	Sum of squares	Df	Mean square	F-Ratio	P-Value
X ₁ :Methanol to oil molar ratio	4356.0	1	4356.0	55.59	0.0000 ^a
X ₂ :Time	441.0	1	441.0	5.63	0.0278 ^a
X ₃ :Catalyst amount	12100.0	1	12100.0	154.43	0.0000 ^a
X ₁ X ₁	2202.7	1	2202.7	28.11	0.0000 ^a
X ₁ X ₂	55.125	1	55.125	0.70	0.4115
X ₁ X ₃	406.125	1	406.125	5.18	0.0339 ^a
X ₂ X ₂	938.08	1	938.08	11.97	0.0025 ^a
X ₂ X ₃	55.125	1	55.125	0.70	0.4115
X ₃ X ₃	5096.54	1	5096.54	65.04	0.0000 ^a
Total error	1567.08	20	78.3542		
Total (corr.)	26301.9	29			

^a P Value less than 0.05 indicates terms are significant.

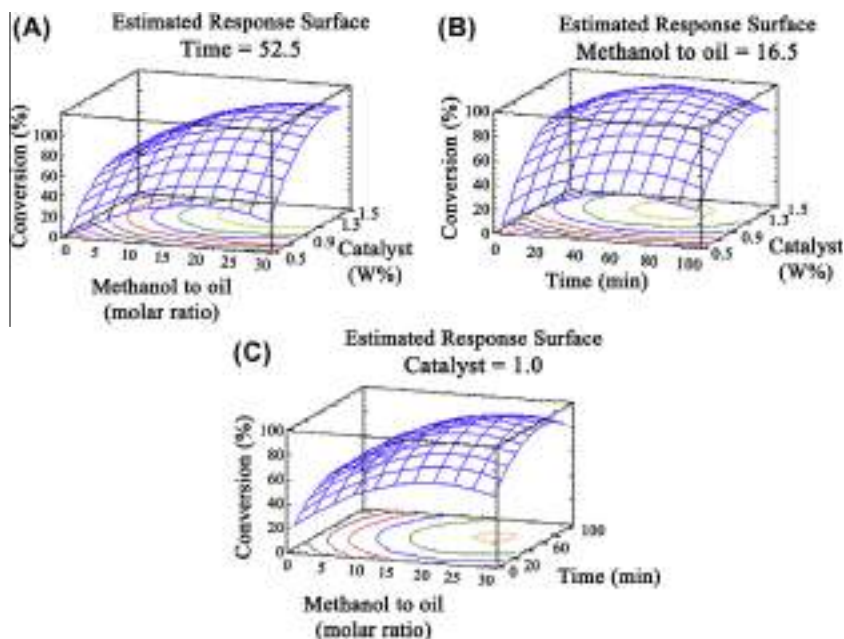


Fig. 1. Response surface plot of combined effects of (a) methanol to oil molar ratios and catalyst amount, (b) time and catalyst amount and (c) methanol to oil molar ratio and time on the conversion percent of transesterification reaction. In each case, other factor is held at zero level.

Table 3

Comparison of the physical properties of the prepared biodiesel with standard biodiesel.^a

Test property	Unit	EN 14212	ASTM D6751	Measured value	Test method
Flash point	°C	120	130	135	ASTM D93
Kinematic viscosity ^b	mm ² /s	3.5–5.0	1.9–6.0	4.874	ASTM D445
Pour point	°C	–	–	0	ASTM D97
Cloud point	°C	–	–	0	ASTM D2500
Density ^c	kg/m ³	860–900	–	896.6	ASTM D 4052
Sulfur content	%	–	0.05 max	<0.01	ASTM D4294
Acid value	mg KOH/g	0.5 max	0.8 max	0.3	ASTM D664

^a The chemical composition was determined by GC-mass as methyl esters of palmitic, 17%, linoleic, 51%, oleic, 20%, stearic, 8%, and linolenic, 4%.

^b Determined at 40 °C.

^c Determined at 15 °C.

Table 4

Comparison of the transesterification activity of the Sr₃Al₂O₆ with some reported basic catalysts.

Conditions and methyl ester content	Sr/ZrO ₂	Sr(NO ₃) ₂ /ZnO	Sr ₃ Al ₂ O ₆	MgAl hydrotalcites	CaO–Al ₂ O ₃
Catalyst loading (wt.%)	2.7	5	1.3	2.5	3.5
Methanol:oil molar ratio	29:1	12:1	25:1	12:1	12:1
Reaction temperature (°C)	115.5	65	60	65	65
Reaction time (min)	87	300	61	240	300
ME content (wt.%)	79.9	94.7	95.7	97.1	94

3.3. Catalyst recycling studies and stability

In order to ascertain whether the catalysts were behaving in a truly heterogeneous manner, the solid catalyst was recovered by centrifugation and separation from reaction mixture after completion of the first run and reusability of the catalyst was investigated in another run by addition to fresh soybean oil and methanol similar to the initial reaction. A decrease in catalyst activity from 95.7% to the 91% was found. The conversion to biodiesel were found to remain at about 83% and 78% as the used catalyst was repeatedly provided to the third and fourth runs, respectively (Fig. S5 in Supplementary). The slight decrease in catalyst activity in the second cycle may have been the effect of the liberated glycerol on the nanocomposite. As seen in Fig. S5, some decrease is observed in biodiesel production after third and fourth cycles. Notably, it was

found that calcination of the used catalyst at above 700 °C prior to the third and fourth cycles exhibits the similar efficiency of the second cycle. The amount of strontium in the fresh and used catalysts were 54.86% and 53.95%, respectively. As indicated in Fig. S6, the EDS of Sr and Al contents in catalyst before and after use are similar. Observation of no catalytic activity of the filtrate of a cycle as catalyst in another cycle in transesterification of soybean oil revealed that no significant desorption has occurred during the course of reaction.

The fuel properties of the prepared biodiesel at the optimal process conditions have been found to comply with the ASTM and EN standards and are presented in Table 3. As seen, the produced biodiesel using Sr₃Al₂O₆ catalyst meets ASTM and EN standard limits.

Due to the presence of Sr, the highly basic Sr₃Al₂O₆ is more active than those of calcium and magnesium containing catalysts

in biodiesel production processes. Compared to the $\text{CaO}/\text{Al}_2\text{O}_3$ catalyst with basicity of $194 \mu\text{mol/g CO}_2$ which has afforded 94% biodiesel, $\text{Sr}_3\text{Al}_2\text{O}_6$ with $1615 \mu\text{mol/g CO}_2$ was found to be a highly basic nanocomposite. For example, utilization of 1.3% $\text{Sr}_3\text{Al}_2\text{O}_6$ in this work within reaction time of 61 min in comparison to other heterogeneous catalysts such as MgAl hydrotalcite and $\text{CaO}/\text{Al}_2\text{O}_3$ respectively with the amounts of 2.5% and 3.5% during reaction times of 4 and 5 h in a highly efficient biodiesel production reaction is promising (Zabeti et al., 2009; Gomes et al., 2011). As indicated in Table 4, $\text{Sr}_3\text{Al}_2\text{O}_6$ exhibits more effective behavior with respect of the other Sr containing catalysts, accordingly (Omar and Amin, 2011; Yang and Xie, 2007).

4. Conclusion

Sr–Al mixed oxides nanocomposite, prepared by sol–gel method showed excellent catalytic activity for transesterification of soybean oil to biodiesel. Results obtained from a Box–Behnken center–united design of response surface methodology (RSM) revealed that individual as well as interaction between variables significantly affect the biodiesel yield. With this information in hand, it was found that utilization of methanol to oil molar ratio 25, reaction time 61 min with 1.3% catalyst loading gave an optimum biodiesel yield of $95.7 \pm 0.5\%$.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.biortech.2013.12.014>.

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