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Research Article

Taguchi Orthogonal Array Based Parameter Optimization of Biodiesel Production from Fish Oil using Ultrasonic Energy

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Abstract: The aim of this study is to investigate the application of ultrasonic energy-assisted biodiesel production process from fish oil catalyzed by KOH at different conditions. The optimization of reaction parameters, such as molar ratio of methanol to oil, catalyst concentration and reaction time, on the transesterification for the production of fish oil methyl ester has been studied. The Taguchi method with an L 9 orthogonal array was implemented to optimize the reaction parameters. The optimal experimental conditions obtained from this study were molar ratio of 9:1, catalyst concentration of 1.5% by Wt. and a reaction time of 30 min. According to Taguchi method, the molar ratio contributed the most important role in the yield of fish oil methyl ester. Validations of the model were done by conducting laboratory experiments. Ultrasonic energy assisted biodiesel production process was proved to be an energy efficient and economically feasible process to produce biodiesel.

Keywords: Biodiesel, fish oil, Taguchi method, transesterification

INTRODUCTION

The growing environmental concerns, the accelerating and frequently fluctuating price of conventional diesel, has sparked renewed attention on the search for an alternative fuel. An alternate to petroleum fuel, biodiesel has emerged as a fuel due to its energy security and lower emissions. Alex and Megan (2008) Biodiesel is one of the widely used renewable energy sources for diesel engines. Biodiesel produced from edible or non-edible oil or from animal fats are potential substitutes for petroleum based diesel fuels. Compared with conventional diesel, biodiesel has the advantages of non-toxic, biodegradable, relatively less inflammable fuel and has significantly lower emissions. Biodiesel is currently produced from edible fatty oils derived from rapeseed, soybean, palm, sunflower, coconut, linseed, animal fats (tallow, lard, poultry fat, fish oils, etc.), or waste cooking oils from the food industry, restaurants or domestic kitchens etc (Eevera *et al*., 2009; Banapurmath *et al*., 2008). Biodiesel produced from edible oil in India is not feasible because of the big gap in demand and supply. To overcome this problem, it has been considered to use other inexpensive, non-edible feedstock for the production of biodiesel (Banapurmath *et al*., 2008). The use of in expensive, non-edible feedstock and the utilization of their byproducts in the biodiesel production may considerably reduce the cost of biodiesel (Shiwu *et al*., 2009). The present research work emphasizes the use of biodiesel produced from fish oil as a suitable alternative for petroleum diesel (Rasim, 2011; Otto and Jan-Erik, 2010; Preto *et al*., 2008; Cherng-Yuan and Rong-Ji, 2009).

India has a vast coastline of 8118 kms and the estimated sustainable resource potential in the marine sector is 3.9 million tons of fish per annum. Total landing of catch (trash) fish is estimated to be around 1.3 million tons, annually (Chandrapal, 2005). In recent years with the fast growth of technology and the commercialization of deep sea fishing activity, the marine fish landing has increased considerably (FAO, 1986). In fish landing almost 40% are considered as trash fish, also the fish processing industry generates large quantities of wastes and by-products which tend to be either discarded or retailed at low value for fertilizer or animal feed (Immanuel, 1996). The present study is the extension and consolidation work based on the previous research results (Rasim, 2011; Otto and Jan-Erik, 2010; Preto *et al*., 2008; Cherng-Yuan and Rong-Ji, 2009).

The most common techniques used for biodiesel production are mechanical stirring and magnetic stirring with a stirring speed of above 300 rpm. Besides, ultrasonic energy can also be used for the reaction. The transient impulsive collapse of the cavitation bubbles disrupts the phase boundary and cause emulsification by ultrasonic jets that impinge one liquid to other Stavarache *et al*. (2005), Mason and Lorimer (1988) and Singh *et al*. (2007), enhancing reaction kinetics.

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Table 1: Characterization of fish oil

Property	Values
Density @15 (°C) (kg/m³)	930
Specific gravity @15 (°C)	0.93
Kinematic viscosity @ 40c in (cSt)	22.90
Flash point (°C)	207
Acid value (mg of KOH/goil)	1.60
Saponifcation value (mg of KOH/goil)	191.30
Iodine value (mgKOH/gmoil)	69.70
Molecular weight (g/mol)	881

Considering these principles, the interfacial region between the oil and alcohol intensively increases, resulting in faster reaction kinetics and higher conversion of oil and product yield. The present research work emphasis the effect of ultrasonic energy on transesterification of fish oil under base-catalyzed transesrerification by varying the process parameters namely the molar ratio, catalyst concentration, the reaction time and the reaction temperature and to optimize the above mentioned reaction parameters. The Taguchi approach with a set of orthogonal arrays was adopted for experimental design, thus the optimal condition of reaction parameters was systematically estimated from the results of nine experimental runs, which had the equivalent effects of 81 experimental runs.

MATERIALS AND METHODS

Materials: Fish oil was commercially purchased from Fish processing unit in Tamil Nadu with the characteristics reported in Table 1. Chemicals such as Potassium Hydroxide Methanol and Anhydrous Sodium sulphate used in the experiments were of analytical grade. Refined Fish oil if purchased from fish processing unit can be used directly without any further purification process. The equipment used for the experimentations are Ultrasonic Bath Sonicator of frequency 33±3 Hz.

Transesterification reaction setup: The reaction was carried out in a 250 mL round bottom flask equipped with a reflux condenser which was immersed in an ultrasonic cleaning bath operating at 33±3 Hz frequency. The reflux condenser was fitted to the neck of the flask. Methanol has a boiling point of 65°C which vaporizes at elevated temperature. The function of the condenser is to condense the methanol that vaporizes during the reaction and to reflux it back into the flask. The bath was filled with distilled water up to 1/3 of its volume (about 2.5 L).

Transesterifcation of fish oil to biodiesel: The pre-treated fish oil was transesterified by varying methanol and catalyst ratios at 65°C using ultrasonic bath. The denser glycerol layer moved to the bottom and the methyl ester layer was separated by allowing it to settle for volume gently until the layer was clear. Vigorous shaking with water was avoided as it would lead to emulsification of methyl esters. Anhydrous sodium sulphate was used to dry the product. The transesterification was done by changing the reaction parameters such as molar ratio, catalyst concentration and reaction time. The optimum value of each parameter involved in the process was determined while the rest of them remain constant. After each optimum was obtained, this value was considered to be constant during the optimization of the next parameter. The percentage of Methyl Ester yield was calculated as follows:

$$\text{Yield of FAME (\%)} = \frac{\text{Weight of Ester (g)}}{\text{Weight of Oil (g)}} \times 100$$

RESULTS AND DISCUSSION

The various results obtained through experimental and theoretical study were discussed as below detailed.

Effect of methanol to fish oil ratio: The molar ratio of alcohol to fish oil is one of the most important variables affecting the FAME yield as well as the production cost of biodiesel. The stochiometric transesterification requires 3 moles of the alcohol per mole of the triglyceride to yield 3 moles of the fatty acid esters and 1 mol of the glycerol (Sinha *et al*., 2008; Leung *et al*., 2010). However, the transesterification reaction is an equilibrium reaction in which a large excess of alcohol is required to drive the reaction close to completion in a forward direction. The effect of methanol to oil ratio was investigated at five different ratios (3:1, 6:1, 9:1 and 12:1, 15:1). The reaction was performed by varying the catalyst concentration and by maintaining a constant reaction time of 30 min. When methanol was added according to the stoichiometry of the reaction as 3:1 molar ratio of methanol to fish oil, the FAME yield was 80%. The FAME yield is increased significantly to 97% in 9:1 molar ratio of methanol to fish oil. However the FAME content decreased with the increase in Methanol addition. The effect of molar ratio on FAME yield is shown in Fig. 1.

Effect of catalyst concentration: Catalyst plays an important role in transesterification reaction. The effect of catalyst concentration was investigated at five different values (1.0, 1.25, 1.5, 1.75 and 2.0% by wt. KOH, respectively). The reaction was performed by varying the methanol to oil ratio and by maintaining a constant reaction time of 30 min. The effect of the catalyst concentration KOH is shown in Fig. 2. It was observed that the excess addition of KOH increased the FAME yield. However the maximum yield of 97% was obtained at a catalyst concentration of 1.5% by wt. of KOH. Increasing the catalyst concentration beyond 1.5% by wt. resulted in decreased FAME yield resulting in soap formation which prevents FAME separation. Several researchers found that catalyst concentration

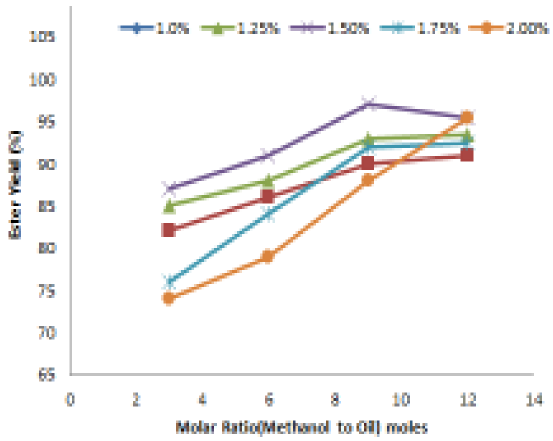

Fig. 1: Effect of molar ratio (methanol to oil) on ester yield at different KOH concentration

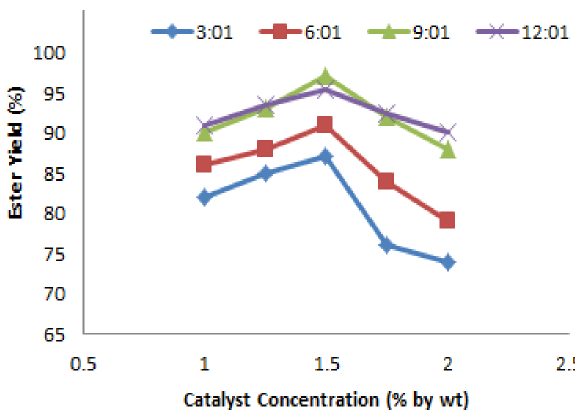

Fig. 2: Effect of KOH concentration on ester yield at different molar ratio of alcohol to oil

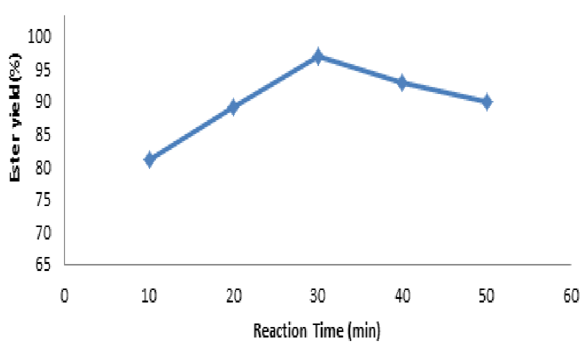

Fig. 3: Effect of reaction time on ester yield

under ultrasonic assisted transesterifcation is usually more than that of homogenous catalysts which does not affect the economics of the process significantly because of simple separation (Venkanna *et al*., 2009; Salamatinia *et al*., 2010).

Effect of reaction time: The reaction time of transesterifcation of fish oil assisted by ultrasonic energy is considerably reduced when compared with conventional process. This is due to reaction speed, increased by ultrasonic cavitational mixing which increases the speed of the reaction by reducing the reaction time. The reaction time is varied from 10 to 50 min in the interval of 10 min, by maintaining a constant methanol/oil molar ratio 9:1 and catalyst concentration of 1.5% by wt. of KOH at temperature 65°C, respectively. The reaction started very fast and almost 81% of the conversion was recorded in the first 10 min. It has been observed that the FAME yield increases with the increase in reaction time. The maximum yield of 97% was obtained at a reaction time of 30 min. The effect of reaction time on the FAME yield is shown in Fig. 3. The increase in reaction temperature speeds up the reaction rate and reduces the reaction time.

Properties of biodiesel: The properties of biodiesel produced from Fish oil was compared with standard Petro-diesel as per ASTM standards (Table 2). The direct usage of fish oil in diesel engine as a fuel was restricted because of the much higher values of fuel properties. Transesterifcation of fish oil into FAME resulted in reduced kinematic viscosity which was within permissible limit. The flash point, fire point and cetane number were in a limit of safe storage and handling Conditions.

Taguchi method for the optimization of transesterifcation of fish oil: Taguchi approach is a statistical technique which uses a set of orthogonal arrays from design of experiments theory to study a large number of variables with a small number of experiments. The orthogonal arrays significantly reduce the number of experimental runs to be carryout. Furthermore, the conclusions drawn from small scale experimental runs are valid over the entire Experimental region spanned by the control factors and their settings (Roy, 2001). Orthogonal arrays are

Table 2: Comparison of properties of biodiesel (produced) and the biodiesel (standard)

Properties	Unit specifications	Specification (biodiesel) BIS India	Specification ASTM D6751	Fish oil biodiesel B100	Diesel
Density @ 15°C	(kg/m³)	860-900	-	880	830
Kinematic viscosity @ 40°C	(cSt)	3.5-5	1.9-6	4.60	2.28
Flash point	(°C)	>100	>130	165	65.50
Cetane number		>51	47	50	45
Fire point	(°C)	-	-	176	70.50
Calorific value	(MJ/kg)	-	-	42.17	44.99

Table 3: Process parameters and their levels

Process parameter	Unit	Symbol	Level 1	Level 2	Level 3
Molar ratio (methanol/oil)	Moles	A	6	9	12
Catalyst concentration	(% by Wt)	B	1.25	1.50	1.75
Reaction time	Min	C	20	30	40

Table 4: Orthogonal array L_9 (3^3) of the experimental runs

Experimental runs	Process parameters and their levels A	B	C	Yield of FAME (%)
1.	1	3	3	87.45
2.	2	2	3	92.58
3.	2	1	2	93.25
4.	1	1	1	81.69
5.	3	1	3	91.86
6.	2	3	1	90.08
7.	1	2	2	91.15
8.	3	3	2	92.38
9.	3	2	1	89.03

Table 5: Yield of methyl esters and S/N ratio

Experimental run	Yield of FAME (%)	S/N ratio
1.	87.45	38.83520
2.	92.58	39.33034
3.	93.25	39.39298
4.	81.69	38.24338
5.	91.86	39.26253
6.	90.08	39.09257
7.	91.15	39.19513
8.	92.38	39.31156
9.	89.03	38.99073

Table 6: Mean S/N ratio for the reaction parameters

Level	Parameter A	Parameter B	Parameter C
1	38.75790330	38.96629667	38.77556
2	39.27196333	39.17206667	39.29989
3	39.18827333	39.07977667	39.14269

Table 7: Results of the ANOVA

Symbol	d.f.	S.S.	M.S.	Contribution (%)
A	2	46.040	23.302	44.64
B	2	5.924	2.962	5.67
C	2	44.696	22.348	42.81
Error	2	6.917	3.458	6.62
Total	8	104.140		

d.f.: Degree of freedom; S.S.: Sum of square; M.S.: Mean square

not unique to Taguchi, they were discovered considerably earlier.

The Taguchi approach of experimental design methodology was employed in this study, with the orthogonal array design used to analyze the effects of the reaction parameters, such as molar ratio, catalyst concentration and reaction time, on the production of fish oil methyl esters. The main operational parameters and levels were based on the experiments conducted and from previously reported studies (Eevera *et al*., 2009; Venkanna *et al*., 2009). The three selected reaction parameters at three levels for this experiment are shown in Table 3. A three level three factor, L_9 (3^3) nine experiments was chosen to perform the experimental runs is shown in Table 4.

The yields of fish oil methyl ester, as biodiesel, prepared under nine sets of experimental conditions are shown in Table 4. The experimental results indicate that experiment run 3 had a maximum yield of fish oil methyl ester of 93.25%, among the nine set of experimental runs with optimal parameters. Experimental run 4 indicates the lowest yield of fish oil methyl ester, at 81.69%. However, it is likely that this would not be preferred way of selecting the optimal conditions using the Taguchi method for the design of an experiment. Taguchi method uses the Signal-to-Noise (S/N) ratio, which are log functions of desired output and serve as objective functions for optimization, helps in data analysis and prediction of optimum results. The S/N ratios are different in terms of their characteristics, of which there are generally three types, i.e., smaller-the-better, larger-the-better and nominal-the-better. The loss function of the higher-the-better, the Mean Squared Deviations (MSD) of each experiment can be expressed by the following equation:

$$MSD = \frac{1}{r}\sum_{i=1}^{r}\left(\frac{1}{y_i}\right)^2$$

where,

r = The number of replications of each experimental run

y_i = The yield of fish oil methyl ester

Then, the S/N ratio was evaluated using the following equation:

$$\frac{S}{N}\ ratio = -10\log(MSD)$$

The S/N ratios for the nine sets of experiments are also shown in Table 5. Experiment run no. 3 indicates the highest yield of fish oil methyl ester 93.25% and had the highest S/N ratio 39.39298. The relationship between the yield of fish oil methyl ester and the S/N ratio was also similarly observed in other experiments. The mean values of S/N ratio at low, medium and high levels for each reaction parameters are shown in Table 6. The reaction parameters which are making an influence in the yield of FAME in terms of S/N ratio are shown in Fig. 4. Based on the S/N ratio results the reaction parameters with highest FAME yield are molar ratio at level 2, catalyst concentration at level 2 and reaction time at level 2, respectively.

Analysis of Variance (ANOVA): The purpose of the ANOVA is to investigate which reaction parameter has significantly affected the yield of fish oil methyl ester. Table 7 shows the summary of ANOVA results indicating the percentage contributions of the reaction

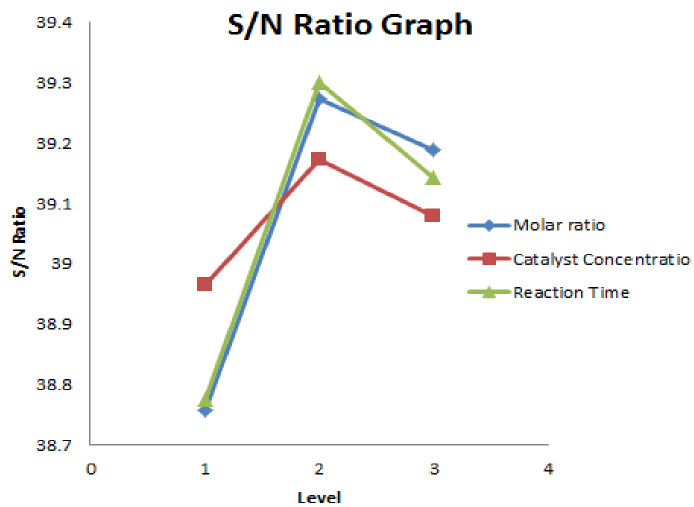

Fig. 4: S/N ratio graph for reaction parameters

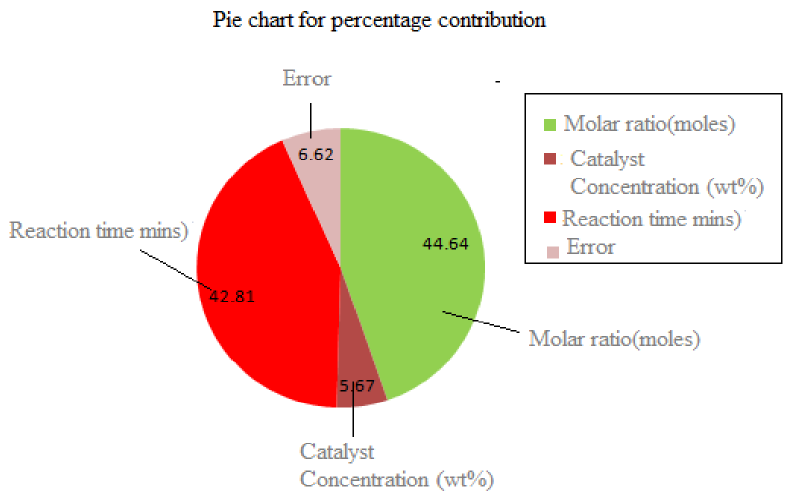

Fig. 5: Percentage contribution of reaction parameters

parameters which makes an influence in the yield of FAME. The ANOVA results indicate that the contribution of reaction parameter molar ratio is more when compared other reaction parameters. The different percentage contribution of reaction parameters are shown in Fig. 5.

From the fig it is clear that molar ratio is the most dominating factor followed by reaction time and catalyst concentration. In order to confirm the optimal conditions derived from this study, experiments were conducted for the production of fish oil methyl ester. The experiments for the optimal reaction parameters, i.e., Molar Ratio at level 2, Catalyst Concentration at level 2 and Reaction Time at level 2, are conducted to obtain the optimal yield of Fish oil Methyl ester of 98.5%. This result was found to be high compared to the result obtained from experiment run no. 3 (93.25%), which gave the highest yield of Fish oil methyl ester. This result showed the significant enhancement of the process performance with the optimized parameters for the production of Fish oil methyl ester.

CONCLUSION

This study dealt with biodiesel produced by transesterification from Fish oil assisted by ultrasonic energy. It was observed that sonication helps the synthesis of methyl esters (biodiesel) from fish oil and higher biodiesel conversion can be obtained within few minutes, whereas the conventional heating process takes more than 60 min. Biodiesel produced by sonication satisfies the ASTM and BIS biodiesel standards. The Taguchi method was employed to design experimental trials, with an ANOVA performed to analyze the relative importance of each experimental parameter on the yield of fish oil methyl ester. The molar ratio, catalyst concentration and reaction time were found to be significant parameters affecting the production of fish oil methyl ester. The contribution of the molar ratio on the production process was found to be the larger than that of any other reaction parameters. The yield of fish oil methyl ester obtained with the optimal experimental parameters was greater than that obtained from experimental run no. 3, which results highest yield from the experimental runs and the theoretically expected value. The experiments conducted under the optimized conditions showed a meaningful enhanced process performance. The Taguchi method provides a systematic and efficient mathematical approach to evaluate and optimize the reaction parameters for the production of fish oil methyl ester.

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