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Biodiesel synthesis from *Styrax officinalis* L. seed oil as a novel and potential non-edible feedstock: A parametric optimization study through the Taguchi technique

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ABSTRACT

The development of renewable and sustainable candidates for petroleum-based fuels is needed to address the present issue of the increasing fuel crisis regarding transportation, environmental pollution, and consumption of the petroleum reserves. In addition, there is a need to explore new non-edible oils for biodiesel production due to problems such as fuel versus food as well as procurement and presence. Therefore the experimental research was carried out to synthesize biodiesel from *Styrax officinalis* L. oil. It has been firstly investigated and reported as a novel feedstock for the production of alternative fuel. The raw material was subjected to esterification at desired reaction parameters estimated by 9-runs (L9) orthogonal approach of Taguchi technique. Noteworthy, the oil content was found to be at 48.29 ± 3.81%. The maximum biodiesel yield of 89.23% was obtained under the following optimized conditions: catalyst concentration of 0.6 wt%, methanol to oil molar ratio of 6:1, reaction duration of 60 min and reaction temperature of 60 °C. The detection of the substantial parameters was achieved using the Taguchi method and the significant parameters were obtained as follows: catalyst concentration, methanol to oil molar ratio, reaction duration, and reaction temperature with contribution factors of 78.07%, 20.32%, 0.42%, and 1.19%, respectively. Statistical analysis employing ANOVA exhibited that emerged outcomes are in good agreement with the predicted values. The fuel properties of the methyl esters from Styrax officinalis L. oil were within the ranges of the EN 14214 specifications. Therefore, the novel seeds can be a suitable feedstock for biodiesel production in the nearest future.

1. Introduction

The energy requirement has been significantly increasing all over the world because of boosting in the industrialization and population. The energy has been considerably consumed by the internal combustion engines, especially diesel engines (compression ignition engines) [1]. Recently, diesel engines are the most common internal combustion engine available in the transportation sector worldwide [2]. The utilization of petroleum-based fuels in the internal combustion engines has been generating a major amount of pollutants throughout the combustion process due to their chemical structure. Therefore, there has been great destruction in the environment as a result of the burning of these fuels [3]. Therefore, researchers have recently begun to focus on the utilization of biodiesel fuels instead of petroleum-based diesel fuel. The demand to look for an alternative to diesel fuel has been further enhanced by the reduction of fossil fuel reserves and the threat of dependence on foreign energy resources [4]. In this context, biodiesel has achieved noticeable ground in the world [5].

Biodiesel is an alternative, renewable, sustainable, clean and important energy source for the diesel engine applications. It has been considered as a non-toxic, biodegradable and environmentally-friendly fuel. In addition, biodiesel has not sulfur content and has lower emission capabilities when combusted in the engines [6,7]. Biodiesel can be synthesized from vegetable oils, animal fats or algae oils by applying different techniques such as dilution, transesterification, thermal cracking (pyrolysis) and microemulsions [8]. Among the above-mentioned techniques, the transesterification method has been mostly preferred technique to obtain biodiesel fuel where the triglycerides react with the short-chain alcohol in the presence of a catalyst [9].

Although it has been reported that there are more than 350 oil-bearing crops as potential raw materials for the biodiesel production in the world, more than 95% of biodiesel has been produced from edible oils such as rapeseed, sunflower, palm, soybean, and coconut oils. The large-scale and continuous production of biodiesel from edible oils has been increasing all over the world. This raises questions of 'Food versus

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Fuel' which is the dilemma concerning the threat of oil crops for biodiesel production to the damage of the food supply [7]. In addition, one of the biggest problems for the production of biodiesel is the cost [10]. On the other hand, the cost of vegetable oils has drastically risen in the last years. For that reason, the economic viability of biodiesel fuels has been influenced significantly [11].

To overcome the above problems, non-edible oils should be evaluated as potential feedstocks for the production of biodiesel. Since edible oils are too expensive to be utilized as a fuel in present days due to the enormous requirement for edible oils as food, the consideration of non-edible oils is very important for developing countries [12]. The most common non-edible feedstocks for the biodiesel industry have been regarded as *Jatropha curcas* [13], *Cerbera odollam* [14], *Calophyllum inophyllum* [15], *Pongamia glabra* [16], *Sapindus mukorossi* [17], *Moringa oleifera* [18], *Croton megalocarpus* [19], *Azadirachta indica* [20], etc. Apart from the above-mentioned non-edible oils, *Styrax officinalis* L. seed oil is a novel and important alternative non-edible raw material for the biodiesel production. The comprehensive information concerning the Styrax officinalis L. seed oil and its biodiesel has been presented in the present study.

1.1. Literature survey

Transesterification reaction heavily depends upon the type of feedstock, water content and free fatty acid (FFA) values of the used oil, catalyst formulation and concentration, alcohol type and alcohol to oil molar ratio, reaction temperature, reaction time, and rotation speed [21]. The process parameters of transesterification reaction can be optimized by various techniques such as conventional [16], response surface methodology (RSM) [22], artificial neural network (ANN) [23], adaptive neuro-fuzzy inference system (ANFIS) [24], and the Taguchi approach [25]. A concise review of parametric study of transesterification variables for biodiesel production is mentioned below.

Kim et al. [26] adopted the Taguchi experimental design to achieve the optimal reaction conditions from the transesterification process of rapeseed oil. They pointed out that the optimized reaction conditions were found to be as follows: KOH concentration of 1.5 wt%, and reaction temperature of 60 °C. Thereby the rapeseed oil methyl ester yield was improved up to 96.7%. Karabas [27] optimized the reaction parameters of biodiesel production from crude acorn (*Quercus frainetto* L.) kernel oil via L9 orthogonal array of the Taguchi technique in order to decline the experimental trials instead of performing 81 experiments. The researcher indicated that the methyl ester yield was 90% under the KOH concentration of 0.7 wt%, methanol to oil molar ratio of 8:1, reaction temperature of 50 °C, and reaction time of 40 min. Another experimental study was conducted by Karabas [28], who applied the Taguchi method for the optimization of reaction parameters on the safflower seed oil biodiesel production. While the yield of biodiesel was found to be at 98%, the effective process parameters on the reaction were determined as alcohol to oil molar ratio. Kumar et al. [29] studied the optimization of transesterification process parameters for the *Manilkara zapota* L. biodiesel applying the Taguchi approach. They achieved the maximum biodiesel yield as 94.83% at 50 °C temperature of reaction, 90 min of reaction time, 6:1 molar ratio of methanol to oil, and 1 wt% of catalyst concentration. Saravanakumar et al. [30] optimized the biodiesel production parameters of Pongamia oil using Taguchi's technique. The optimum reaction parameters giving the maximum biodiesel yield of approximately 86% were found to be as 550 rpm of stirrer speed, 15 g of NaOH catalyst, and 80 min. of reaction time. Kumar et al. [31] used the Taguchi method with an L9 orthogonal array in order to analyze the effect of factors on the safflower oil transesterification parameters. The results demonstrated that the biodiesel yield was 93.8% with a 4:1 of methanol to oil molar ratio, 1.5 wt % catalyst concentration, 90 min of reaction time and 60 °C of reaction temperature. In addition, the catalyst concentration was the most efficient parameters on the yield of biodiesel with the contribution factor of 51.1%. Gadhave and Ragit [32] carried out the *Vernicia fordii* oil biodiesel by applying L9 orthogonal array matrix of the Taguchi method and they noticed that the yield of biodiesel was enhanced up to 93.20% under the optimum reaction conditions. Also, the methanol to oil molar ratio was found to be the most influential parameter on the biodiesel yield. Interestingly, Senthilkumar et al. [33] compared the Taguchi and Box-Behnken methods for the production of biodiesel from non-edible wild radish oil and remarked that the first experimental design exhibited almost similar results of Box-Behnken within the limited number of tests. Karmakar et al. [34] concluded that the maximum FFA conversion was obtained as 90.83% from castor oil and the contributing factors of the significant parameters were calculated as follows: oil to methanol molar ratio (59.6%), agitation speed (16.95%), reaction temperature (12.59%), and catalyst concentration (10.82%), respectively. Dhawane et al. [35] examined a statistical optimization work of biodiesel synthesized from waste cooking oil via Taguchi L9 orthogonal array matrix. The contributing factors of reaction temperature, methanol to oil molar ratio and reaction time were found to be in the order of 71.6%, 21.5%, and 5.3%.

1.2. Botanical description and distribution of Styrax officinalis L.

The scientific name "*Styrax officinalis* L." which is a shrub plant that grows in Turkey's South East, Mediterranean, Aegean, Marmara and Middle Black Sea regions. It can be seen that it also grows in biomes in Southeast Asia, Mediterranean and tropical parts of America. The systematic position of *Styrax officinalis* L. is known as follows: Plantae – Plants (Kingdom), Tracheobionta – Vascular plants (Subkingdom), Spermatophyta – Seed plants (Superdivision), Magnoliophyta – Flowering plants (Division), Magnoliopsida – Dicotyledons (Class), Dilleniidae (Subclass), Ebenales (Order), Styracaceae – Storax family (Family), *Styrax* L. – Snowbell (Genus), *Styrax officinalis* L. (Species) [36]. In addition, Fig. 1 shows the distribution of *Styrax officinalis* L. plant around the world.

Styrax officinalis L. can grow widely between 0 and 1000 m altitudes. It is an alternative oilseed plant in the form of perennial shrubs. Styrax is known to have 8 genera and about 130 species [38,39]. In addition, the existence of this plant has been known since ancient times. Strabon, the famous geographer of ancient times, when talking about Selge, it is known that the gives information about the rich vegetation of the region as well as the styrax tree in his book of "Geographika".

The high oil content (approximately 50%) in its seeds indicates that the plant can provide significant ecological and economic benefits if cultivated within the scope of agricultural forestry. This plant is more valuable in the industrial terms because it can grow in non-agricultural areas, barren and damaged surfaces, forests and even in shades of 60–70%. Moreover, it has very suitable features because of "new plants in the new climatic conditions" discussing the concepts of global warming and climate change [40]. However, it has not been sufficiently taken into account. The utilization of oil in the seeds as an energy resource in the industrial application can decline the demand pressure on the edible oils. At the same time, it can contribute to supply food security all over the world [41–43].

The plant, flower, and fruits of the *Styrax officinalis* L. were illustrated in Fig. 2. These photographs were taken in the fields during the observations from authors. The local name of this plant is "Tesbi" or "Tesbi çalısı" in Turkey because the rosary is made from hard and bead-like seeds of the fruit. Furthermore, it is also seen that this name varies according to regions. For instance, "Tesbi" has been used as a name of this plant around Antalya and Isparta provinces in Turkey while it is known as "Zanzalak" around Mersin. As a result of field research, we detected that people also entitled of this plant as "Ayı fındığı (in Turkish)-bear nuts" or "Yaban ayvası (in Turkish)-wild quince".

Oflas [44] reported that there were 65 plants in 150.00 m^2 after the preliminary survey. It can be considered that there may be 4300 plants in 1.00 ha area with a ratio calculation according to the aforementioned evaluation. In a preliminary study we conducted, approximately 9.00 kg of the *Styrax officinalis* L. seeds were harvested from the point of



Fig. 1. The distribution of *Styrax officinalis* L. plant around the world [37].

729 m altitude (02.95964 east, 41.16141 north) of the Amanos Mountains within the boundaries of Nurdağı district of Gaziantep province. From these values, 4300.00 plants × 9.00 kg seeds = 38700.00 kg seeds can be harvested from 1 ha forest area. If it is assumed that the fruit peel is 30.00% of the total fruit weight, 25800.00 kg of seed remains. Internal rate of seed to total seed is approximately 30.00% and this weighs 7740.00 kg. Since the seed has approximately 50.00% of oil, 3870.00 kg of vegetable oil can be obtained from 1.00 ha. This rate is the average oil yield of many plant under cultivation in Turkey represents a value on that too. Taking into

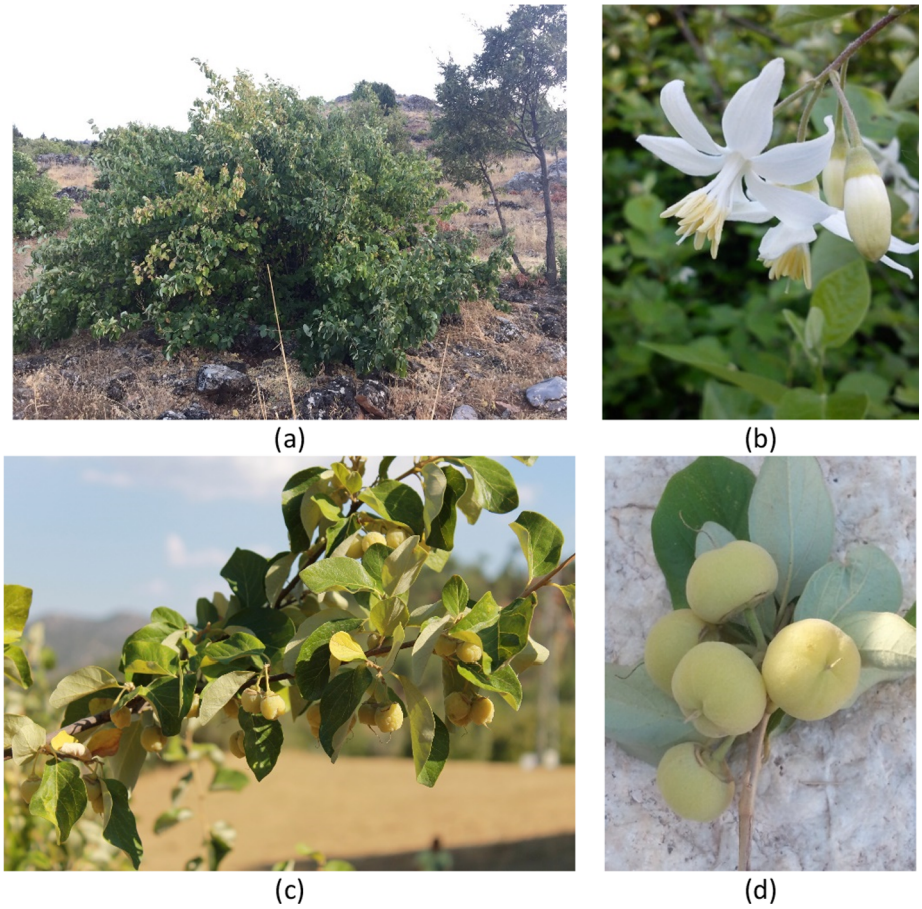


Fig. 2. (a) The plant, (b) flower, and (c, d) fruits of the *Styrax officinalis* L.

account the harvest, transportation and processing of 38700.00 kg biomass, which has the potential to be obtained from an area of one hectare, it is obvious that it will make a significant contribution to employment. If 3637.80 kg of 38700.00 kg biomass is separated as oil, 35062.20 kg biomass is left behind. Considering the processes of using the crust portion of this biomass as fuel, different forest products and pulp, a serious economic benefit will occur. This biomass, which can be obtained from the plant, does not have the potential to be obtained from many cultivated plants.

Overall evaluation, *Styrax officinalis* L. seed, and its oil should be considered as a major alternative energy resource for the biodiesel industry. However, as far as we know, the production of biodiesel from *Styrax officinalis* L. seed oil has not been reported from the researchers. Therefore, it is the first implementation related to the *Styrax officinalis* L. seed oil biodiesel. With this experimental study, a novel and important potential feedstock will be recommended for biodiesel production.

1.3. Scope of the study

The detail literature survey demonstrated that the oilseeds could be evaluated for the production of biodiesel taking into account of their physicochemical properties. Furthermore, the presentation of novel feedstocks with beneficial characteristics for the production of biodiesel fuel could have a substantial influence on the biodiesel industry. In addition, the progress of new raw materials is able to lead to the conservation of the most resources all over the world. The main objective of the present study is to show the *Styrax officinalis* L. seed oil could be utilized as a novel and potential non-edible feedstock for the production of biodiesel. To the best of the authors' knowledge, it has not yet been reported that the biodiesel has been obtained from *Styrax officinalis* L. seed oil. Therefore, it is the first attempt to investigate the optimization of the most important reaction parameters such as methanol to oil molar ratio, catalyst concentration, reaction temperature and reaction time for the methanolysis of *Styrax officinalis* L. seed oil. The statistical optimization study was performed with the L9 orthogonal array of the Taguchi's method in order to improve the yield of biodiesel. Moreover, the physicochemical properties of *Styrax officinalis* L. seed oil methyl ester were investigated and compared with the global biodiesel standards of EN 14214 and ASTM D6751.

2. Materials and methods

2.1. Materials and reagents

Styrax officinalis L. fruits were collected from nature in three different places in Göksu Valley in Mut, where is a district of Mersin in Turkey's Mediterranean region. The properties of collection places are as follows: 36°48′ 4″ N-33° 8′ 16″ E (The altitude is 420 m), 36° 47′ 55″ N-33° 6″ 43″ E (The altitude is 770 m), and 36°47″ 58″ N-33°6′ 13″ E (The altitude is 860 m). The sampling sites were shown in Fig. 3. The authentication of the plant was realized at Yozgat Bozok University, Yozgat, Turkey. In addition, it would be an appropriate case to give the environmental conditions of the collection place. Within the average of 1927–2018, the average temperature, average maximum temperature, average minimum temperature, average sunshine duration, average rainy days, and average precipitation per year were found to be as 19.2 °C, 23.3 °C, 14.8 °C, 90.1 h, 66.6 days, and 597.3 mm [45] respectively. The ripe *Styrax officinalis* L. fruits were picked in the first week of September 2018.

The seeds of the *Styrax officinalis* L. were separated from fruits manually and the outer shell was cracked with the help of suitable types of equipment. After removing the fruit peel, the inner oilseeds were cleaned prior to the oil extraction process. Furthermore, the seeds were dried in the sunlight for a week in order to decrease the moisture ratio. The appearances of fruits and seeds were presented in Fig. 4.

The chemicals and other materials were chosen on the foundation of simplicity and ease of obtainment. In the transesterification reaction, methanol and NaOH were selected as a short-chain alcohol and base catalyst due to the reaction activity. Methanol (99.8%) and diethyl ether (> 99.5%) were supplied from Isolab (Wertheim, Germany). Sodium hydroxide (NaOH) pellet (99%) and petroleum-benzene solvent were purchased from Merck Chemical Company (Darmstadt, Germany). Ethanol (> 99.5%) and reference standards of the fatty acid methyl esters (> 99%) were procured from Sigma-Aldrich Chemical Company (St. Louis, Missouri, USA), phenolphthalein indicator and KOH solution (0.1 N) were utilized from Norateks Chemical Company (Istanbul, Turkey). The qualitative filter paper (125 mm) was supplied from S&H Labware (Ankara, Turkey). All of the reagents were used as a received form not to apply further purifications because of analytical grade. The distilled water was produced by using Millipore brand Direct-Q 8UV model.

2.2. Extraction of crude oil from Styrax officinalis L. seeds

The crude oil of *Styrax officinalis* L. seeds was obtained with the help of a screw press and heating for the production of biodiesel. On the other hand, soxhlet extraction equipment (Foss brand Soxtec 2055 model) was used in order to determine the exact oil ratio of the seeds. The measuring range, reproducibility, temperature range of the soxhlet extraction equipment are 0.1–100%, ± 1% relative and 0–285 °C, respectively. The accuracy of this equipment was found to be at ± 1%. Shivani et al. [46] conducted an experimental study to extract Jatropha seed oil from its seeds with the help of various techniques (separating funnel, centrifugation, filtration and soxhlet extraction). The soxhlet extraction is a better technique for obtaining crude oil from the seeds as compared to the above-mentioned methods due to the fact that this method is a continuous and there is a complete recovery. Samuel and Emovon [47] investigated the effect of extraction process parameters such as solvent to seed ratio (from 2 mL/g to 6 mL/g) and time of the extraction (from 1 h to 8 h) on the yield of *Pycnanthus Angolensis* seed oil which is a novel potential biofuel candidate using response surface methodology (RSM). As a result, the highest yield of *Pycnanthus Angolensis* seed oil was found to be at 38.64% at the following optimal level of 6 mL/g of solvent to seed ratio and 8 h for the duration of the extraction. The extraction procedures outlined by Waheed et al. [48] was adopted. The *Styrax officinalis* L. seeds were separated from foreign matters. The dried seeds were crushed to the fine particles with a hand mill in order to obtain large surface area of the cells containing oil for the oi extraction process. The *Styrax officinalis* L. seed oil was initially extracted from the collected seeds in the laboratory conditions because of the unavailability of the *Styrax officinalis* L. seed oil in the local markets. 2 g of seeds extracted thoroughly using a petroleum-benzene solvent in a soxhlet extractor in three times. The extraction yield of the feedstock was found according to the following equation:

$$Oil\ content\,(\%) = \frac{M_{oil\ produced}}{M_{sample\ used}} x100 \quad (1)$$

The *Styrax officinalis* L. seed crude oil was settled in a sealed glass bottle for two weeks so as to subside the pulp particles with the aid of density difference and gravity effect. The upper oil phase was discharged into the other similar bottle. Then the filtering process was applied to the crude oil in order to eliminate the suspended pulp particles by using cloth material. Next, the qualitative filter paper was used for the advanced filtering process. Fig. 5 shows the *Styrax officinalis* L. seed crude oil. Consequently, the filtered oil experimented in biodiesel production.

2.3. Experimental apparatus for the biodiesel production

In the present study, the single-step transesterification process was performed in order to produce biodiesel from the Styrax officinalis L.

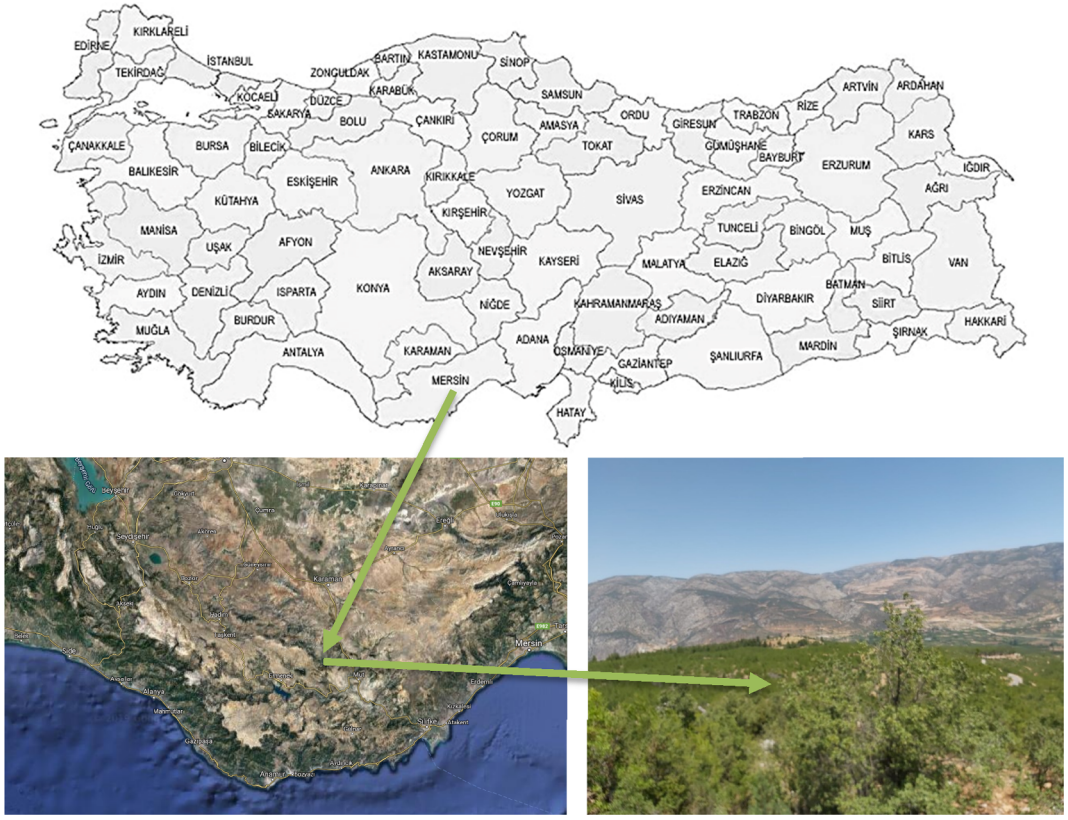


Fig. 3. Geographic locations and the collection place of the *Styrax officinalis* L. in Mut, Mersin, Turkey.

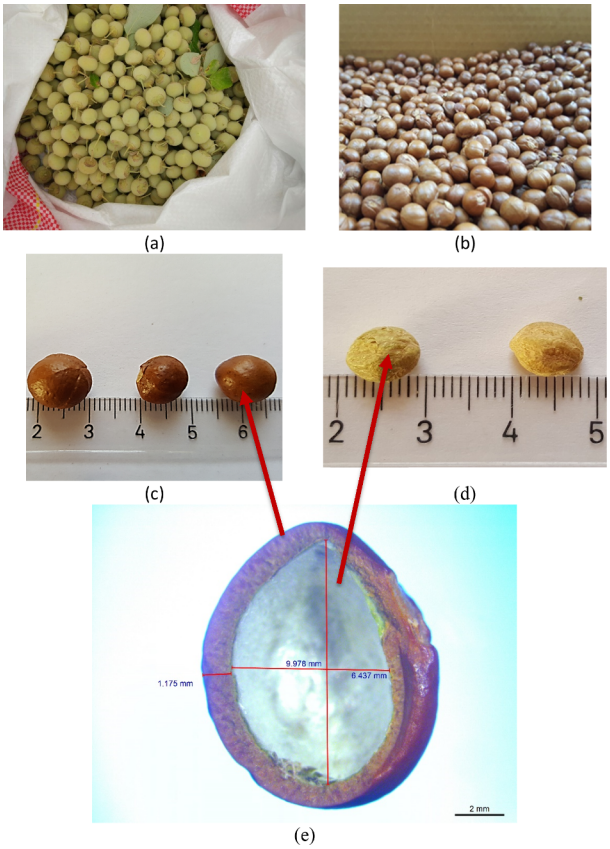


Fig. 4. The appearances of fruits and seeds: (a) The ripe fruits, (b) cleaned seeds, (c) dimension of the outer shell, (d) dimension of the inner seeds, (e) microscopic view of the seed.



Fig. 5. The *Styrax officinalis* L. seed crude oil.

seed oil. For this purpose, a small-scale biodiesel reactor was used. The aforementioned reactor was consisted of a 250 mL three-necked and flat bottom flask fitted with a reflux condenser to avoid the evaporation of alcohol, placed on the magnetic stirrer with hot plate (Scilogex brand MS7-H550-Pro model), sampling outlet, and condensation systems. The experimental setup was presented in Fig. 6.

2.4. Transesterification process of Styrax officinalis L. seed oil

It has been well known that one of the most important key properties of feedstocks is FFA content due to specifying regarding the kind of the transesterification method such as a single-step or double-step

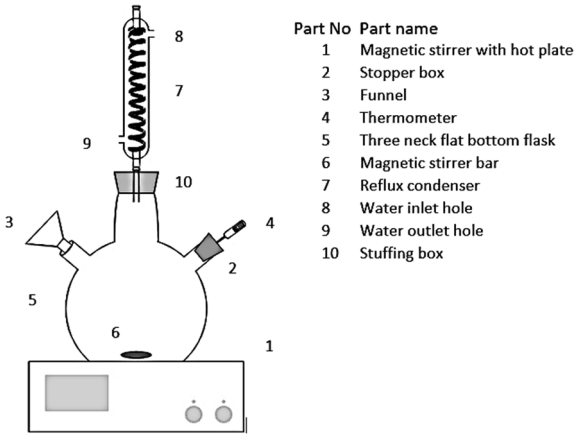


Fig. 6. Experimental setup for producing methyl ester.

transesterification process. Although many researchers have given different opinions concerning the limit value of FFA for determining the step size of the transesterification reaction with a base catalyst, the general acceptance is that FFA should be lower than 2% [49–51]. If the FFA content of the feedstock exceeds 2%, the two-step transesterification process should be preferred. In the present study, FFA content was measured by applying the acid-base titration technique. It can be concluded that a single-step transesterification process with NaOH catalyst has been implemented owing to the fact that FFA content of *Styrax officinalis* L. seed oil was found to be at 1.94%.

The catalyst concentration, methanol to oil molar ratio, time of the reaction and temperature of the reaction have been accepted as the most significant parameters that affect the biodiesel production by most of the investigators [52–54]. In the present work, above-mentioned parameters were considered for the optimization in the transesterification process in order to produce *Styrax officinalis* L. seed oil biodiesel with the maximum percentage of yield. Therefore, the selected levels for these parameters were given in Table 1. The factor levels of the studied variables were selected in the account of the operating limits of the biodiesel yield and the conditions of the biodiesel production process of the individual variables that had an impact on the preliminary tests.

The single-step transesterification process has been briefly summarized as follows: First of all, 100 g (± 0.01) of *Styrax officinalis* L. seed oil was poured into the three-necked flat-bottom batch reactor and the temperature was increased to reach the desired level. The stirrer speed was kept constant at 1000 rpm in all trials. The methanol-catalyst mixture was prepared by dissolving the re-measured amount of base catalyst (NaOH) inside the required concentration of alcohol. When the temperature of the *Styrax officinalis* L. seed oil was reached to the desired level, the obtained methoxide solution was slowly added into the reactor with the aid of a funnel. After completion of the pouring process, the reaction time was initiated by using a chronometer. The reflux condenser was mounted on top of the three necks in order to avoid evaporation and reuse the alcohol. Fig. 7 illustrates the transesterification reaction.

The mixture in the reactor was poured into a 250 mL separation conical funnel at the end of the predetermined time of the reaction. After 8 h of settling, the bottom layer (glycerol) was received from the separation funnel with the help of the drainage valve. Then, the crude biodiesel produced from *Styrax officinalis* L. seed oil was poured in a glass beaker and reheated up to 75 °C in order to remove the excess methanol in biodiesel. Next, the crude biodiesel was taken into the separation funnel and cooled until the temperature reached to 55 °C. It was gently washed with distilled water at the same temperature so as to eliminate impurities, unreacted catalyst, and methanol. The wastewater was removed from the funnel with the help of the valve and washed biodiesel was heated till the temperature reached the 120 °C in order to dewatering process. Lastly, the biodiesel was stored in a glass bottle on the dark medium and rested for decreasing the temperature. Upon reaching the ambient temperature, the samples were measured with the aid of a precision scale. The percentage yield of the biodiesel was calculated using the following equation:

$$Biodieselyield\,(\%)\text{: } Y = \frac{gramsofproducedbiodiesel}{gramsofusedoilinthereaction} x100 \quad (2)$$

The process flow chart of the production of biodiesel from the *Styrax officinalis* L. seed crude oil via transesterification method was illustrated in Fig. 8.

After the biodiesel was produced from *Styrax officinalis* L. seed oil under the optimal reaction conditions, the physicochemical properties were analyzed. ASTM and EN test methods were followed in order to measure the properties and the results were compared with the global biodiesel standard of the EN 14214.

2.5. Experimental design using orthogonal array

The Taguchi method introduced into the literature by Dr. Genichi Taguchi has not investigated every probable combination of the process parameters. However, this technique has reduced the bulky optimization procedure to the just a number of combinations. In general, the Taguchi approach has gained attention from the researchers in industrial applications as well as in the biodiesel production. A minimum number of trials has been required, which facilitates the collection of data, reducing valuable time and resources in order to light of process factors with a substantial effect on the product yield. In this context, the most important parameter that affects the yield of biodiesel could be predicted according to the proportion of influence. In the present research, the L9 orthogonal array was preferred using the relation of Eq. (3) and conducted owing to considering the number of parameters and their levels. The statistical analysis of the Taguchi design of experiments was carried out using Minitab 16 software.

$$N = (L - 1)P + 1 \quad (3)$$

where N is defined as the smallest possible number of runs, P is the number of parameters and L is the number of levels.

2.6. Selection of the control parameters and their levels

The transesterification process was influenced a lot of parameters such as alcohol type and alcohol to oil molar ratio, catalyst type and catalyst concentration, time of the reaction, temperature of the reaction, quality of the reagents, FFA and water contents of the feedstock, stirring or agitation speed. Among them, only the four most important and effective parameters were considered in this work at three levels (P = 4 and L = 3 as presented in Table 1). The minimum experimental runs were calculated using Eq. (2) and the necessitate number of the test was found to be at 9 as per L9 orthogonal array matrix with four parameters at three levels (3^4) exhibited in Table 2. If the Taguchi method has not been implemented in this investigation, the experiments were carried out at 81 different combination of reaction conditions. Unfortunately, each experiment has not been adopted thrice due to the lack of feedstock.

Table 1
The reaction parameters and their levels.

Reaction parameters (unit)		Levels		
		1	2	3
A	Catalyst concentration (wt. %)	0.6	0.9	1.2
B	Methanol to oil molar ratio (mol/mol)	4	6	8
C	Time of the reaction (min)	60	90	120
D	Temperature of the reaction (°C)	40	50	60

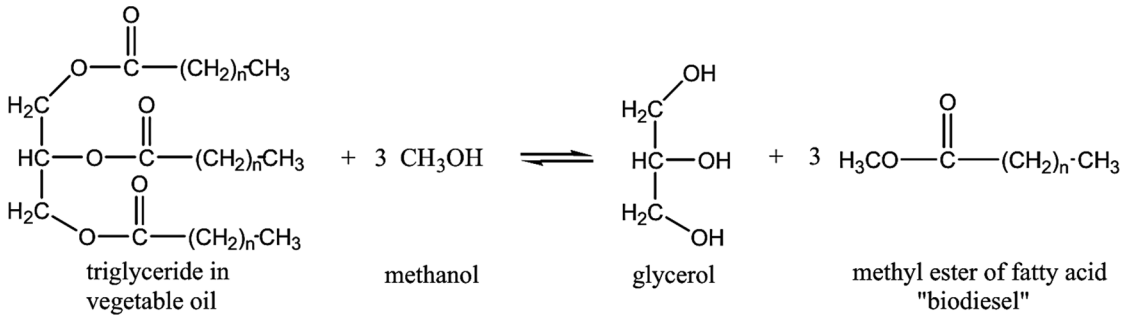


Fig. 7. Transesterification reaction [29].

2.7. Signal to noise ratio (SNR) and analysis of variance (ANOVA)

The Taguchi technique supplies the calculation of signal to noise ratio (SNR) based on the results acquired from the runs. SNR is described as the ratio of the average of the biodiesel yield to the standard deviation. The SNR can be used efficiently to achieve the prime level of each parameter as well as the associated optimum conditions to maximize the efficiency of the process. In this context, there are several types of SNR preferred in the Taguchi method in the point of the consequence of the issues. They could be indicated as follows: LTB-larger the better, STB-smaller the better, and NTB-nominal the best and the SNR (dB) models were tabulated in Table 3.

In the present study, the aim is to improve the yield of biodiesel from the *Styrax officinalis* L. seed oil, thus, the LTB model was selected. On the other hand, SNR cannot identify the factor that substantially affects output and the degree to which each factor contributes to the output. For this reason, it is able to understand by predicting the

Table 2
L9 orthogonal array fro design of experiment.

Experiment no	Parameters (unit) and their levels			
	Catalyst concentration (wt. %)	Methanol to oil molar ratio (mol/mol)	Time of the reaction (min)	Temperature of the reaction (°C)
1	1	1	1	1
2	1	2	2	2
3	1	3	3	3
4	2	1	2	3
5	2	2	3	1
6	2	3	1	2
7	3	1	3	2
8	3	2	1	3
9	3	3	2	1

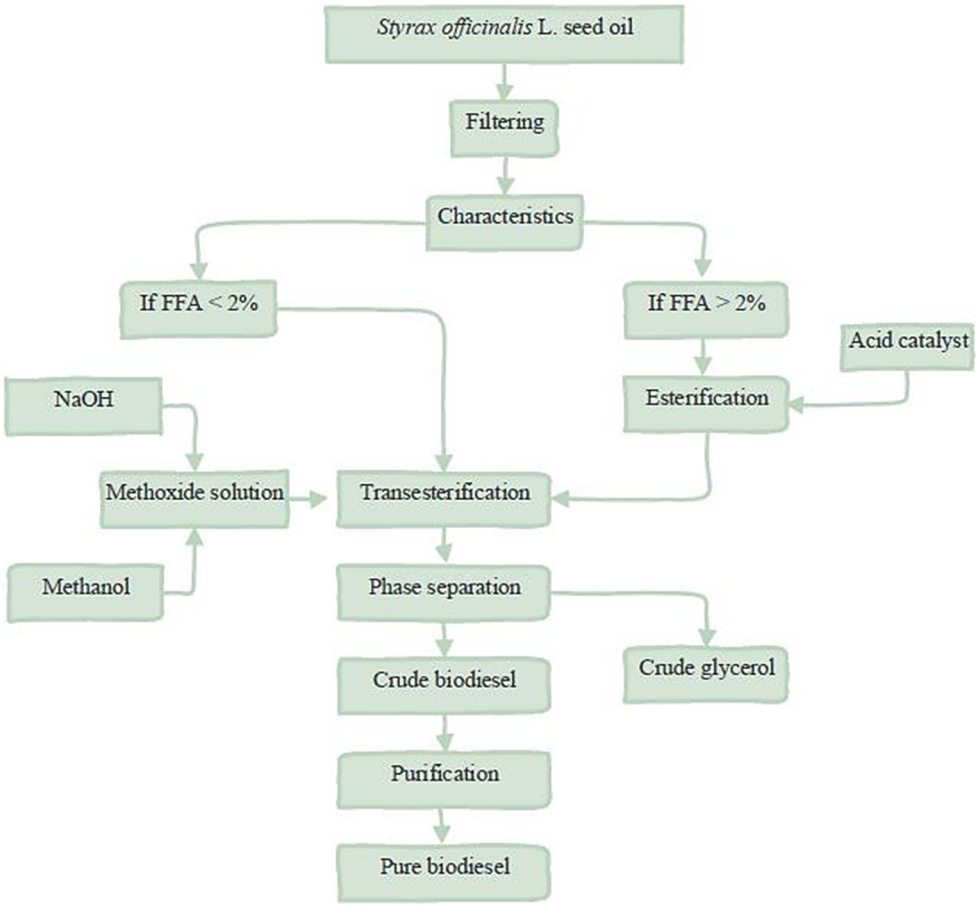


Fig. 8. The process flow chart of the production of biodiesel from the *Styrax officinalis* L. seed crude oil.

Table 3
Signal to noise ratio (SNR).

SNR	SNR calculation formula
Larger the better	$SNR_i = -10x\log_{10}\left(\sum_{j=1}^{N}\frac{\left(1/y_j^2\right)}{n}\right)$ (4)
Smaller the better	$SNR_i = -10x\log_{10}\left(\sum_{j=1}^{N}\left(y_j^2\right)/n\right)$ (5)
Nominal the best	$SNR_i = 10x\log_{10}\left(\bar{y}_i^2/s_i^2\right)$ (6)

where
$\bar{y}_i = \frac{1}{n}\left(\sum_{j=1}^{n} y_{i,j}\right)$ (7)
$s_i^2 = \frac{1}{n-1}\left(\sum_{j=1}^{n} y_{i,j} - \bar{y}_i\right)$ (8)
$\bar{y}_i$: the mean value of response.
s_i^2: the variance.
i: the experiment number.
j: the trial number.
n: the number of trials.

statistical indicators via the analysis of variance (ANOVA). One of the most important sign is the contribution factor of the process parameters which can give an opinion concerning their influence on the process. The contribution factors of each parameter have been calculated by the following formula:

$$\%\ contribution\ factor = \frac{SS_f}{SS_T} \times 100 \quad (9)$$

where SS_f is the sum of the squares for the f_{th} factor and SS_T is the total sum of squares of all parameters. SS_f and SS_T could be computed using Eq. (10) and Eq. (11), respectively.

$$SS_f = \sum_{j=1}^{3} n[(SNR_L)_{fj} - SNR_T]^2 \quad (10)$$

$$SS_T = \sum_{i=1}^{9} (SNR_i - SNR_T)^2 \quad (11)$$

where SNR_L is the level mean signal to noise ratio, n is the number of experiments at level 'j' of factor 'f'.

2.8. GC–MS analysis of oil and biodiesel

The fatty acid composition of the *Styrax officinalis* L. seed oil and its biodiesel product were determined by using Shimadzu brand QP2010 model (Kyoto, Japan) Gas Chromatograph (GC) system equipped with DB-5MS capillary column (30 m × 0.32 mm × 0.25 μm) and compared with the most common feedstocks used in the production of biodiesel. The analysis status was presented as follows: after the sample was injected, the temperature was augmented up to 70 °C for 1 min at the beginning of the process. Then 120 °C at 20 °C/min (held for 2 min), 180 °C at 10 °C/min (held for 3 min), lastly 240 °C at 5 °C/min (held for 10 min). The other conditions are adjusted as the injector temperature-250 °C, split ratio (1:10), carrier gas (helium), gas flow rate (1.5 mL/min), ionization mode used at electronic impact (70 eV), and the volume of injected sample (1 μL).

2.9. Physicochemical properties of Styrax officinalis L. seed oil and biodiesel

The main physicochemical properties of *Styrax officinalis* L. seed oil and the some of the fuel properties of the produced *Styrax officinalis* L. seed oil under the optimized reaction conditions of the transesterification procedure were determined according to the ASTM D6751 and EN 14214 specifications. Group I metals (Na + K) and Group II metals (Ca + Mg) (EN 14538), density (EN ISO 3675), polyunsaturated (≥4 double bonds) methyl esters (EN 15779), linolenic acid methyl ester (EN 14103), cloud point (ASTM D2500), pour point (ASTM D97), copper strip corrosion (EN ISO 2160), ash content (EN ISO 3987), flash point (ASTM D93), pH and freezing point were detected based on the cited standards. The saponification number, iodine value, cetane number, oxidation stability, kinematic viscosity and higher heating value were calculated with the following Equations 16–21 as per the recommended by several authors.

The acid value (AV) and FFA content were measured thanks to the suitable acid-base titration method with KOH solution and the phenolphthalein indicator and they can be calculated as given by Eq. (12) and Eq. (13) [34,35].

$$AV\,(mgKOH/g) = \frac{(v-b)xNx56.1}{w} \quad (12)$$

$$FFA\,(\%) = \frac{(v-b)xNx28.2}{w} \quad (13)$$

where v is the volume of titer solution, b is the blank volume, w is the mass of the oil, and N = 0.1 is the strength (normality) of the KOH solution. Also, 28.2 indicates the presence of the oleic acid content in the feedstocks while 56.1 denotes the molar weight of the KOH.

The degree of unsaturation (DU) was found using the following empirical equations [55,56]:

$$DU = (monounsaturatedC_n{:}\ 1,\ wt.\ \%) + 2(polyunsaturatedC_n{:}\ 2,\ wt.\ \%) + 3\ (polyunsaturatedC_n{:}\ 3,\ wt.\ \%) \quad (14)$$

Long-chain saturated factor (LCSF) can be predicted from the amount of the saturated fatty acids and their melting points (MP_n) by using the Equation (15) [55,56]:

$$LCSF = \sum\left(\frac{MP_n xC_n}{100}\right) \quad (15)$$

The saponification number (SN), iodine value (IV), cetane number (CN), oxidation stability (OS), kinematic viscosity at 40 °C, and higher heating value (HHV) of the *Styrax officinalis* L. seed oil biodiesel can be calculated as given the following formulas [4,57,58]:

$$SN = \sum\left(\frac{560xA_i}{MW_i}\right) \quad (16)$$

$$IV = \sum(254xDxA_i)/MW_i \quad (17)$$

$$CN = 46.3 + \frac{5458}{SN} - 0.225IV \quad (18)$$

$$OS = -0.0518(DU) + 11.121 \quad (19)$$

$$\ln(\eta_i) = -12.503 + [2.496 \times \ln(MW_i)] - (0.178 \times N_s) \quad (20)$$

$$HHV = 49.43 - [0.041(SN) + 0.015(IV)] \quad (21)$$

where A_i and MW_i are the proportions and molecular mass of each component in the fatty acids, respectively. D denotes the number of double bonds. η_i defines the kinematic viscosity (40 °C), HHV is the higher heating value, and N_s represents the number of double bonds in the monounsaturated and polyunsaturated fatty acids.

Lastly, the molecular weight (MW) of the crude oil obtained from *Styrax officinalis* L. seeds were calculated using Eq. (22) as given below [29]:

$$MW = 3 * (AMW_{FA}) + weight\ of\ glycerol\ backbone \quad (22)$$

where AMW_{FA} represents the average molecular weight of all fatty acids.

It is to be noted that the sample calculation of the aforementioned physicochemical properties of biodiesel produced from the *Styrax officinalis* L. seed oil is presented in the Appendix.

3. Results and discussion

3.1. Styrax officinalis L. seed oil content

Most of the countries have commercially produced biodiesel fuel from the vegetable oils. In general, these oils have been consumed as food by people. In this context, this case has led to the struggle of fuel versus food supply all over the world. Therefore nonedible feedstocks can be evaluated in the production of biodiesel fuel; especially novel raw materials are very significant because the cost of biodiesel is highly depended on the expense of feedstock at ratios between 70 and 85% [59]. Interestingly, the oil content of its seeds was found to be at 48.29 ± 3.81% by the Soxhlet method.

There are a few published articles in the recent literature regarding the oil quantification of this new and potential alternative raw material as far as we know. It has been reported and presented for the first time that *Styrax officinalis* L. seed oil could be utilized as a novel, potential and promising substitute for the production of biodiesel considering the high percentage of oil content as compared to the other feedstocks. Moreover, the comparison of the oil content of *Styrax officinalis* L. seed with other feedstocks commonly used in the biodiesel production was also represented in Fig. 9. It is also validated with the literature survey that the oil content of *Styrax officinalis* L. seed was the highest oil content. The use of edible oils in biodiesel production has been risky in terms of food safety and costly as well [60]. Although food shortages and starvation around the world have been increasing day by day, the countries have been still producing biodiesel from the edible grade oils such as sunflower, canola, palm, etc. It is well known that the deaths from starvation due to food shortages have been realized in low-income countries. Migrant mobility from Asia, Africa, and South America to developed countries is level unstoppable no longer. In this context, it is very important to produce biodiesel from non-edible oils obtained from non-agricultural areas all over the world [61,62]. For this purpose, a large number of wild plants have been investigated in order to find alternative vegetable oil sources that are not agricultural production and suitable for biodiesel transformation. It can be concluded that the cost of *Styrax officinalis* L. production is going to be lower than the other field crops when it is entered in agricultural production [63]. Therefore *Styrax officinalis* L. seed oil should be evaluated in the production of biodiesel due to its non-edible properties. Overall considerations and trials have shown that the *Styrax officinalis* L. seed oil has a promising potential for the production of biodiesel in worldwide avoiding the fuel vs. food.

3.2. The fatty acid composition and physicochemical properties of Styrax officinalis L. seed oil

The crude seed oil from *Styrax officinalis* L. plant has been considered for the production of biodiesel without any further refining process. The fatty acid composition and physicochemical characteristics of the *Styrax officinalis* L. seed oil have been investigated to indicate the compatibility of as a feedstock for the production of biodiesel. In addition, an appropriate production type was chosen according to its FFA content and other properties. Table 4 tabulates the comparison of the fatty acid compositions of the *Styrax officinalis* L. seed oil with various raw materials considered in the biodiesel production by different researchers. The GC analysis graph was also illustrated in Supplementary data file A. The maximum content of unsaturated fatty acid was found to be linoleic acid (64.41%) and the other one was oleic acid (23.22%). Based on Eqs. (14) and (15), DU and LCSF were calculated as 152.86 and 7.67, respectively. These are higher than that of Jatropha and Soybean oils.

Table 5 presents the physicochemical properties of *Styrax officinalis* L. seed oil. Density at 20 °C, 0.909 g/cm^3; FFA content (as oleic acid), %1.94; molecular weight, 876.11 g/mol; acid value, 3.88 mg KOH/g; physical state at room temperature, liquid; color, greenish yellow; odour, agreeable; pH, 6.0; flash point, 228 °C; calorific value, 38.652 MJ/kg; water content, 554.50 ppm.

3.3. The effects of reaction parameters on the biodiesel yield

Fig. 10 (A-F) illustrates contemporaneous influences of catalyst concentration (0.6, 0.9 and 1.2 wt%), methanol to oil molar ratio (4:1, 6:1 and 8:1), time of the reaction (60, 90 and 120 min) and temperature of the reaction (40, 50 and 60 °C) on the percentage biodiesel yield of *Styrax officinalis* L. seed oil. The aim of the present study is to increase the biodiesel yield through the statistical approach. In other words, the maximization of the biodiesel yield is the objective. The yield of

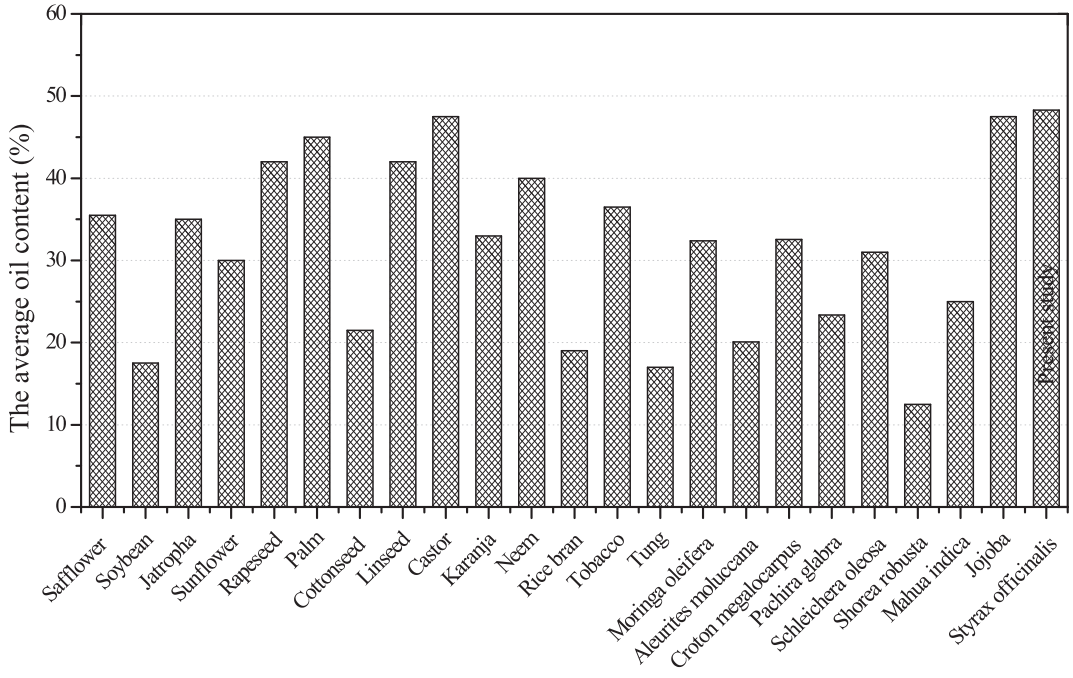


Fig. 9. The comparison of the oil content of *Styrax officinalis* L. seed with other feedstocks commonly used in the biodiesel production [64–70].

Table 4
The comparison of the fatty acid compositions of the *Styrax officinalis* L. seed oil with various raw materials.

No	Fatty acid	Structure	Formula	Molecular weight	Systematic name	*Styrax officinalis* L. (Present study), %	Jatropha oil (%) [56]	Canola oil (%) [71]	Soybean oil (%) [55]
1	Myristic	14:0	$C_{14}H_{28}O_2$	228.38	Tetradecanoic	0.03	3.0	–	–
2	Palmitic	16:0	$C_{16}H_{32}O_2$	256.43	Hexadecanoic	8.08	13.4	3.49	11.3
3	Palmitoleic	16:1	$C_{16}H_{30}O_2$	254.43	Hexadec-9-enoic	0.01	–	–	0.1
4	Margaric	17:0	$C_{17}H_{34}O_2$	270.46	Heptadecanoic	0.10	–	–	–
5	Margaroleic	17:1	$C_{17}H_{32}O_2$	268.46	*cis*-9-heptadecenoic	0.14	–	–	–
6	Stearic	18:0	$C_{18}H_{36}O_2$	284.49	Octadecanoic	3.00	3.6	0.85	3.6
7	Oleic	18:1	$C_{18}H_{34}O_2$	282.49	*cis*-9-Octadecenoic	23.22	51.2	64.4	24.9
8	Linoleic	18:2	$C_{18}H_{32}O_2$	280.49	*cis*-9-*cis*-12 Octadecadienoic	64.41	28.8	22.3	53.0
9	Linolenic	18:3	$C_{18}H_{30}O_2$	278.49	*cis*-9-*cis*-12 Octadecatrienoic	0.10	–	8.23	6.1
10	Arachidic	20:0	$C_{20}H_{40}O_2$	312.54	Eicosanoic	0.07	–	–	0.3
11	Gondoic	20:1	$C_{20}H_{38}O_2$	310.54	11-Eicosenoic	0.12	–	–	0.3
12	Behenic	22:0	$C_{22}H_{44}O_2$	340.59	Docosanoic	0.47	–	–	0
13	Erucic	22:1	$C_{22}H_{42}O_2$	352.62	*cis*-13 Docosenoic	0.25	–	–	0.3
14	Lignoceric	24:0	$C_{24}H_{48}O_2$	368.65	Tetracosanoic	–	–	–	0.1
15	Nervonic	24:1	$C_{24}H_{46}O_2$	366.65	*cis*-15-Tetracosenoic acid	–	–	–	–
Saturated fatty acids						11.75	20.0	4.34	15.3
Monounsaturated fatty acids						23.74	51.2	64.4	25.6
Polyunsaturated fatty acids						64.51	28.8	30.53	59.1
Total fatty acids						100	100	99.27	100
DU						152.86	108.8	–	143.8
LCSF						7.67	6.4	–	1.6

Table 5
Physicochemical properties of *Styrax officinalis* L. seed oil.

No	Properties	Units	Values
1	Density at 20 °C	g/cm^3	0.909
2	Free fatty acid content	%	1.94
3	Acid value	mg KOH/g	3.88
4	Molecular weight	g/mol	876.11
5	Physical state at room temperature	–	Liquid
6	Color	–	Greenish yellow
7	Odour	–	Agreeable
8	pH	–	6.0
9	Flash point	°C	228
10	Calorific value	MJ/kg	38.652
11	Water content	ppm	554.50

biodiesels produced applying lower catalyst concentration (e.g. 0.6 wt %) and moderate methanol to oil molar ratio (e.g. 6:1) possessed the highest values due to the fact that higher catalyst concentration led to decrease the yield of biodiesel because of the tendency of soap formation. When the influence of catalyst concentration on the biodiesel yield took into account, it can be underlined that NaOH amount was the most effective factor. But, further increasing of NaOH from 0.6 wt% caused to decrease in the biodiesel yield. The majority of the researchers pointed out that the optimum catalyst amount was found to be at 0.6 wt %. However, the implementation of basic NaOH catalyst in the transesterification process with higher ratios (e.g. 0.8 wt% and 1.2 wt%) could result in more soap formation in the reaction because of the presence of FFA in the oil. In addition, the separation of glycerol could be difficult and thereby the biodiesel yield was significantly reduced.

Le Chatelier's principle has indicated that the reaction of transesterification is an equilibrium reaction; therefore, it has shifted the methanolysis reaction towards the forward direction. Consequently, an assertive balance for biodiesel formation could be provided for increasing biodiesel yield. The anymore rise in time of the reaction and methanol to oil molar ratio led to rising the glycerol solubility in the samples. This caused the intervention of a small amount of glycerol with the ester phase. Thereby, the glycerol in the ester phase was the result of more foam formation and the ester yield was decreased.

The alterations in the time of the reaction and temperature of the reaction could not substantially affect the variation characteristics of the percentage yield of biodiesel in accordance with the catalyst concentration and methanol to oil molar ratio simultaneously as aforementioned. It means that nearly identical change movement was observed when the time of the reaction and temperature of the reaction were varied as presented in Fig. 10(F). This could be verified with the statistical analysis results. The time of the reaction and temperature of the reaction were found to be least effective parameters than others on the biodiesel yield.

On the other hand, considering the temperature of the reaction, the transesterification reaction has the activation energy, i.e., the reaction is endothermic. Taking into account of reaction temperature, increasing from 40 °C to 60 °C had a positive impact on the transesterification, resulting in improve the biodiesel yield. In other words, the maximum biodiesel yield was found to be at the highest temperature of the reaction. 60 °C was selected as a reaction temperature control parameter because of the boiling point of the used alcohol (64.7 °C). If the reaction temperature is chosen near or above the boiling point of the alcohol, it can cause to encourage the saponification process of the triglycerides in the oil by NaOH prior to the finishing alcoholysis; thus, it led to decrease the ester yield.

3.4. Identification of optimal reaction conditions by the Taguchi approach

The percentage yield of *Styrax officinalis* L. seed oil biodiesel under the planned nine runs of experiments, their SNR values and overall mean SNR_T value were given in Table 6. In the present experimental study, for the maximization of the biodiesel yield produced from a novel feedstock of *Styrax officinalis* L. seed oil, the larger the better (LTB) SNR mode has been used in the software in order to get the suitable structure. The analysis results indicated that the highest biodiesel yield was found to be at 88.30% with the experiment number 2 and the lowest biodiesel yield was determined as 75.43% with the experiment number 7.

Although the arrangement of the parameters corresponds to the experiment number 2 exhibits the maximum biodiesel yield in the trials, this set could not be the optimum reaction parameters for the *Styrax officinalis* L. seed oil. For this reason, the signal to noise ratio level (SNRL) would be evaluated in order to obtain the correct optimum set of reaction parameters. "SNRL is defined as the algebraic mean of all the SNRs of a particular control parameter at a specified level." In this

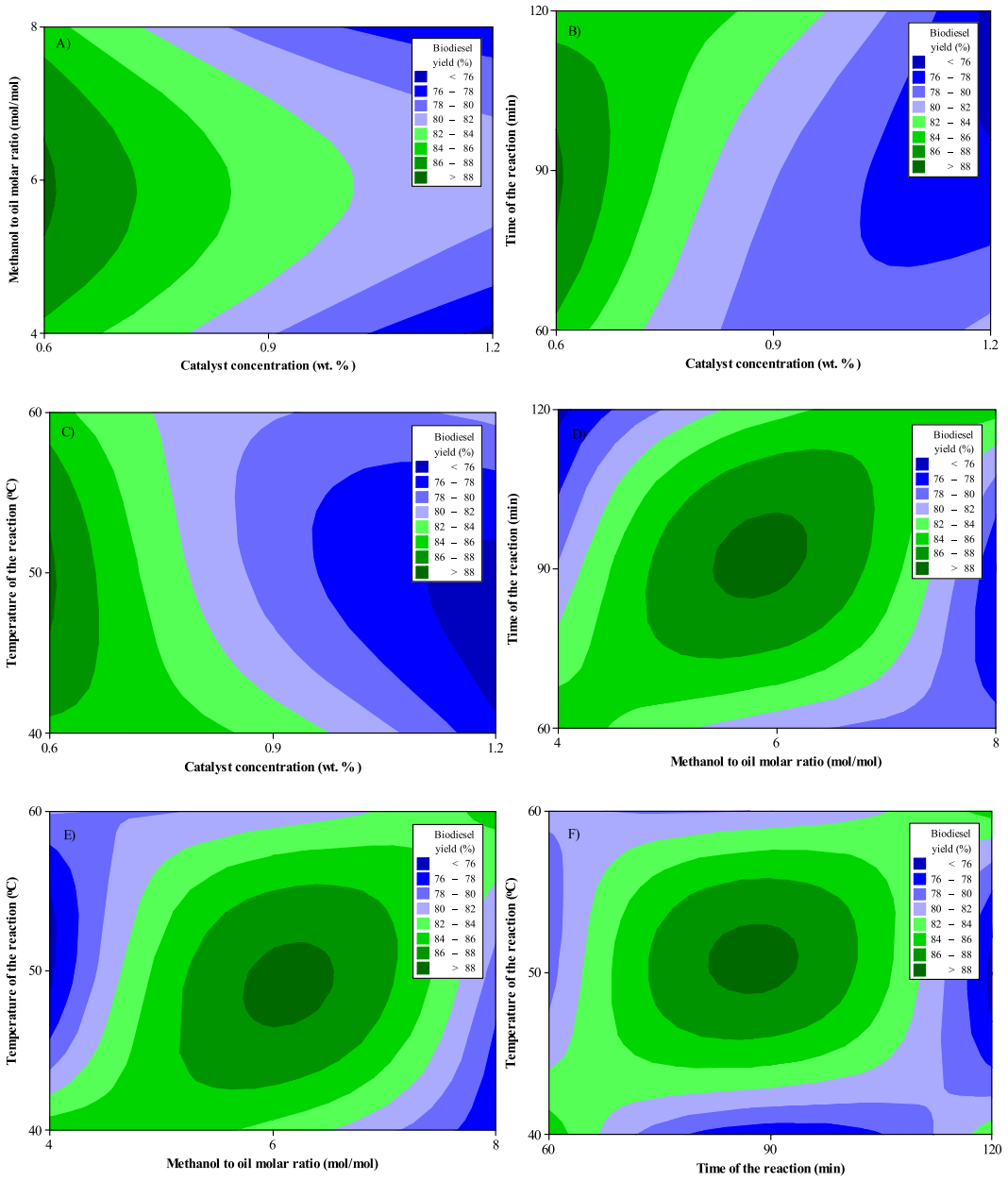


Fig. 10. Contour plot of biodiesel yield (%) in accordance with **A)** catalyst concetration (wt. %) and methanol to oil molar ratio (mol/mol), **B)** catalyst concetration (wt. %) and time of the reaction (min), **C)** catalyst concetration (wt. %) and temperature of the reaction (°C), **D)** methanol to oil molar ratio (mol/mol) and time of the reaction (min), **E)** methanol to oil molar ratio (mol/mol) and temperature of the reaction (°C), **F)** time of the reaction (min) and temperature of the reaction (°C).

Table 6
The biodiesel yield, SNR and SNR_T values for the nine runs of experiments.

Experiment no	A	B	C	D	Biodiesel yield (%)	SNR
1	0.6	4:1	60	40	85.37	38.63
2	0.6	6:1	90	50	88.30	38.92
3	0.6	8:1	120	60	84.73	38.56
4	0.9	4:1	90	60	80.17	38.08
5	0.9	6:1	120	40	83.30	38.41
6	0.9	8:1	60	50	79.17	37.97
7	1.2	4:1	120	50	75.43	37.55
8	1.2	6:1	60	60	80.57	38.12
9	1.2	8:1	90	40	76.43	37.67
SNR_T-Overall mean						38.21

study, for instance, for parameter C (time of the reaction) at level 1, SNRL was computed as 38.24 corresponding to the data (38.63, 37.97, and 38.12) received from in order of experiment numbers 1, 6, and 8; at level 2 SNRL was found to be at 38.22 using the related values (38.92, 38.08, and 37.67) taken from experiment numbers 2, 4, and 9, respectively; finally at level 3 SNRL was calculated as 38.17 using the values (38.56, 38.41, and 37.55) regarding experiment numbers 3, 5, and 7, respectively. SNRL at each level of factors, DSNRL (the difference between maximum and minimum of SNRLs of a particular parameter) and the rank of factors were separately calculated and tabulated in Table 7. The rank was determined according to the values of DSNRL. In other words, the maximum DSNRL was highlighted as a rank 1 while the minimum DSNRL was marked as a rank 4. Thanks to the ranks, the most and least effective parameters on the yield of *Styrax officinalis* L. seed oil biodiesel were identified. When Table 7 was evaluated, catalyst concentration was found to be the most influencing factor on the percentage yield of biodiesel. Methanol to oil molar ratio and the temperature of the reaction were the second and third effective parameters followed by the time of the reaction.

Table 7
SNRL at each level of factors and rank of factors.

Level	SNR corresponding to biodiesel yield			
	A	B	C	D
1	38.70	38.09	38.24	38.23
2	38.15	38.49	38.22	38.15
3	37.78	38.07	38.17	38.25
Delta	0.92	0.42	0.07	0.11
Rank	1	2	4	3

Table 8
The results of ANOVA.

Parameters	Degree of freedom	Sum of squares	Mean square	Percentage contribution (%)
A	2	1.29038	0.645189	78.07
B	2	0.33590	0.167950	20.32
C	2	0.00681	0.003406	0.42
D	2	0.01969	0.009844	1.19
Error	0	–	–	–
Total	8	1.65278	0.826389	100

For better understanding, the effects of each factor at three different levels on the yield of *Styrax officinalis* L. seed oil biodiesel in the point of SNRL are presented in Fig. 11. First of all, the greater value of SNRL means a larger effect of a particular factor at that level. In addition, the highest values of SNRL in each graph pointed out the optimum reaction conditions of those particular factors on the *Styrax officinalis* L. seed oil biodiesel. In this context, the optimum reaction conditions for the maximization of the percentage yield of biodiesel were found to be as A (catalyst concentration) at level 1 (0.6 wt%), B (methanol to oil molar ratio) at level 2 (6:1), C (time of the reaction) at level 1 (60 min), and D (temperature of the reaction) at level 3 (60 °C).

3.5. Analysis of variance (ANOVA)

The most important process parameter has been determined by computing the percentage of each parameter's contribution to the biodiesel yield. The results of the analysis of variance (ANOVA) were tabulated in Table 8. As observed, it could be monitored that the process parameter A (i.e. catalyst concentration) having 78.07% contribution on the yield of *Styrax officinalis* L. seed oil biodiesel was found to be the most substantial factor followed by the process parameter B (i.e. methanol to oil molar ratio) with 20.32% contribution. Among the process parameters, the time of the reaction was observed as the least effective process parameters having 0.42% contribution followed by the temperature of the reaction with a 1.19% contribution. The transition of the methanol to the oil has been slow in the first few minutes of the transesterification reaction. But, after five minutes, it was reported that the reaction was fast [72], i.e. the conversion rate at the beginning of the reaction is high and is not affected after reaching a steady state. It also presents that a large amount of biodiesel conversion reached close to the start of the reaction.

3.6. Prediction of maximum biodiesel yield and its validation

As mentioned above, the theoretical percentage yield of *Styrax officinalis* L. seed oil biodiesel was found to be at 89.23% by means of the Taguchi approach under the optimum reaction conditions. The optimum transesterification reaction parameters were obtained as follows: NaOH concentration of 0.6 wt%, methanol to oil molar ratio of 6:1, time of the reaction of 60 min, and temperature of the reaction of 60 °C. However, these conditions should be experimentally verified. Therefore, the production of biodiesel from the *Styrax officinalis* L. seed oil was carried out experimentally at the optimum reaction conditions as given above.

There is no set of the experiment as shown in Table 6 in order to confirm the theoretical percentage of yield. Therefore, the biodiesel from *Styrax officinalis* L. seed oil was produced thrice via the transesterification reaction at the optimized reaction conditions so as to eliminate the errors. As a result, the yield of biodiesel for trial 1, trial 2, and trial 3 were calculated as 88.64%, 88.12%, and 89.54%, respectively. The average percentage of the three experiments was found to be at 88.77%, providing much the matches with that of theoretically predicted rate. As seen, there is a slight difference between the predicted and experimental values. This could be explained with the effect of the unpredictable variables. The major barriers of the integrated process are the high equipment investment, large energy consumption,

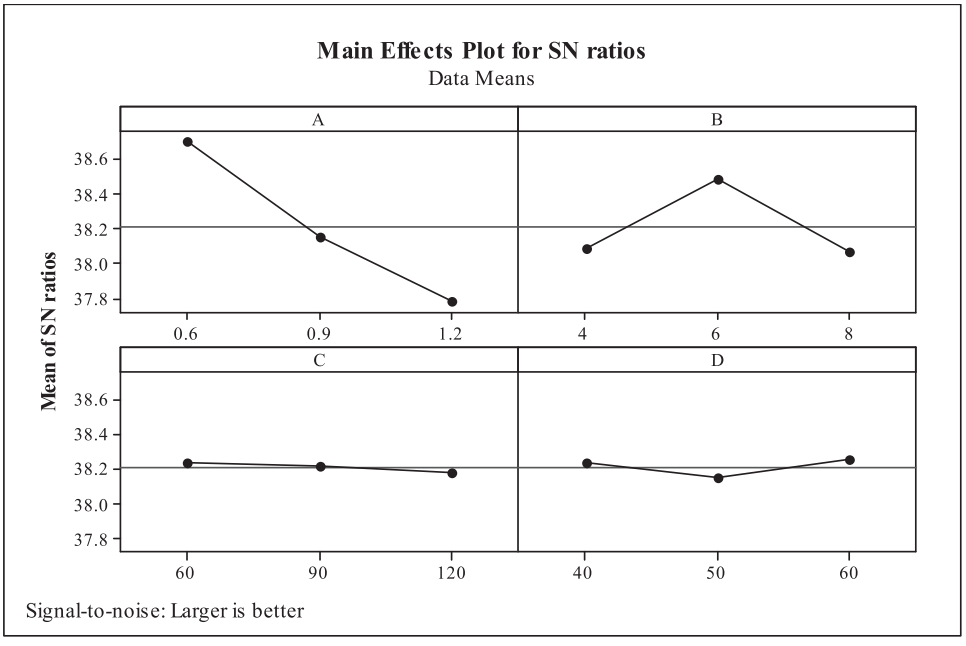


Fig. 11. SNRL of each factor at three different levels.

Table 9
The comparison of reaction parameters of *Styrax officinalis* L. seed oil biodiesel with other less known or novel feedstocks.

Feedstock	Type and volume of reactor (mL), type of agitator	Production process parameters							Optimization technique	Optimal reaction conditions					References
		Catalyst type and amount (wt.% of the oil)		Alcohol type and alcohol to oil molar ratio		Reaction temperature (°C)	Reaction time (min)	Agitation intensity (rpm)		Catalyst amount (wt.% of the oil)	Alcohol to oil molar ratio	Reaction temperature (°C)	Reaction time (min)	Yield (%)	
Styrax officinalis L. seed oil	Three-necked flat-bottom flask, 250, magnetic	NaOH	0.6–1.2	Methanol	4:1–8:1	40–60	60–120	1000	Taguchi, L9	0.6	6:1	60	60	89.23	This work
Idesia polycarpa var. *vestita* fruit oil	Four-necked round bottom flask, 2000, mechanical	KOH	0.5–2.0	Methanol	4.5:1–6.5:1	20–60	20–60	600	Conventionel	1.0	6:1	30	40	> 99	[73]
Calophyllum inophyllum L. seed oil	Three-necked round bottom flask, 500, mechanical	KOH	0.75–1.5	Methanol	4:1–10:1	45–65	30–150	700	Conventionel	1.25	8:1	60	120	89	[74]
Sinapis alba L. seed oil	Three-necked flask	NaOH	0.1–0.9	Methanol	2:1–10:1	50–70	15–75	600	Conventionel	0.5	6:1	65	75	92	[75]
Moringa oleifera seed oil	Three-necked flask, 250, magnetic	Nano-MgO	0.5–1.5	Methanol	6:1–12:1	45–65	120–240	–	Taguchi, L9	1	12:1	45	240	93.69	[76]
Vitis vinifera seed oil	Conical flask	KOH	0.5–1.5	Methanol	4:1–8:1	30–60	60–90	700	Taguchi, L9	1	6:1	60	60	96.90	[77]
Ailanthus altissima seed oil	Ultrasonication device, amplitude:70%, pulse:70	KOH	1–2	Methanol	2:1–12:1	50	2–10	–	Box-Behnken factorial design	1.01	8.5:1	50	4.71	92.26	[78]
Raphanus sativus seed oil	Three-necked round bottom flask, 500, magnetic	KOH	0.5–1.5	Methanol	3:1–9:1	40–60	20–40	500	Taguchi, L9	1	9:1	50	30	94.58	[79]
Quercus frainetto L. kernel oil	A small-scale laboratory rotary evaporator, 1000 mL round flask, electrical mill	KOH	0.5–1.0	Methanol	6:1–10:1	50–55	40–80	600	Taguchi, L9	0.7	8:1	50	40	90	[27]

and high production cost. Table 9 exhibits ranges of the reaction parameters implemented and the highest biodiesel yield achieved in the prior investigations of different base methanolysis of less known or novel oilseed or biodiesel feedstocks using different optimization techniques. For comparison, the reaction conditions and the maximum biodiesel yield obtained from the present study are also presented. It should be noted that there is a lack of study regarding the optimization of transesterification process parameters using novel feedstocks. Therefore it needs to be conducted more research to fill this gap. There is a great deal of work for researchers.

3.7. Physicochemical properties of biodiesel obtained from Styrax officinalis L. seed oil

Some of the major physicochemical properties of biodiesel obtained from *Styrax officinalis* L. seed oil like density, kinematic viscosity, acid value, heating value, iodine value, cetane number, pH, cloud point, pour point, and etc. were determined and reported in Table 10. The fuel properties were also compared with the global biodiesel standards of EN 14214 and ASTM D6751. The findings showed that most of the physicochemical properties of Styrax officinalis L. seed oil methyl ester satisfied the standards and thereby it could be a prospective candidate to the petroleum-based diesel fuel for the diesel engine application.

3.8. Elemental analysis results of Styrax officinalis L. seed oil biodiesel

Fig. 12 presents the elemental analysis results of the *Styrax officinalis* L. seed oil biodiesel. The actual values of the elements were also tabulated in Supplementary data file B. As seen, the contents of Al, Cr, Mn, Co, Ni, and Cu in the biodiesel were found to be lower than 1 ppm. The most abundant element was obtained as Fe with 38.22 ppm following Ca with the value of 35.57 ppm.

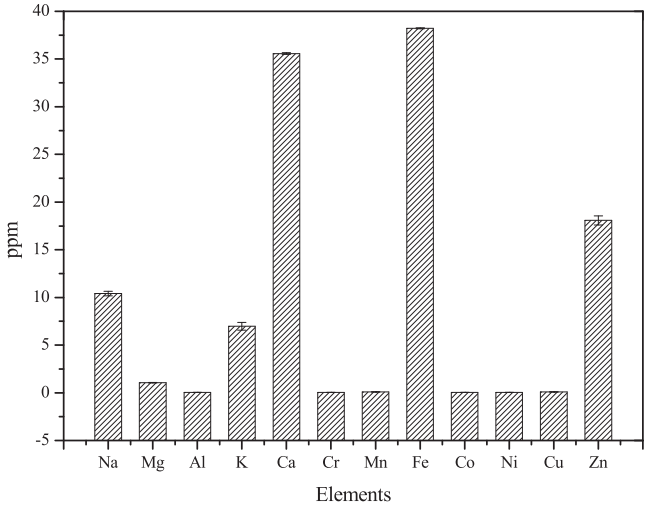


Fig. 12. The elemental analysis results of the *Styrax officinalis* L. seed oil biodiesel.

4. Conclusion

In the present experimental research, it is the first study to investigate the statistical optimization of the most important process parameters such as methanol to oil molar ratio (4:1, 6:1, and 8:1), catalyst concentration (0.6 wt%, 0.9 wt%, and 1.2 wt%), reaction temperature (40 °C, 50 °C, and 60 °C)and reaction time (60 min, 90 min, and 120 min) for the methanolysis of *Styrax officinalis* L. seed oil through Taguchi technique. In other words, *Styrax officinalis* L. seed oil has not been used in the production of biodiesel until this paper and a novel biodiesel synthesized from *Styrax officinalis* L. seed oil has been worked and reported. Interestingly, most of the physical characteristics of the plant and its seeds which are grown in the non-agricultural areas are found to become much more economical than many oil seeds. Based on the results, the following conclusions can be drawn:

- The oil content of *Styrax officinalis* L. seed was measured as 48.29 ± 3.81% and it has a higher oil rate than that of other raw materials used in the biodiesel production. Although it has high oil content in the seeds, interestingly *Styrax officinalis* L. seed oil has not been considered in the biodiesel production till this paper.
- On the other hand, the authors have thought that this attempt is not enough for the literature and further studies should be conducted.

Table 10
Properties of *Styrax officinalis* L. seed oil biodiesel in comparison with EN 14214 and ASTM D6751 biodiesel standards.

No	Fuel properties	Units	ASTM D6751	EN 14214	The present study
1	Density (at 15 °C)	g/cm^3	880	860–900	886[b]
2	Higher heating value	kJ/kg	–	–	39,023
3	Cetane number	–	Min. 47	Min. 51	40.47
4	Kinematic viscosity (at 40 °C)	mm^2/s	1.9–6.0	3.5–5.0	3.57
5	Iodine number	g iodine/100 g	–	Max. 120	147.20
6	Acid value	mg KOH/g	Max. 0.5	Max. 0.5	0.4
7	Cloud point	°C	−3 to −12	–	−2.5
8	Pour point	°C	−15 to −16	–	−9.5
9	Cold filter plugging point	°C	Max. + 5	–	−5.4
10	Flash point	°C	Min. 93	Min. 101	175
11	Copper strip corrosion (3 h at 50 °C)	Degree of corrosion	Class 3	Class 1	1a
12	Carbon	wt. %	77	–	76.91
13	Hydrogen	wt. %	12	–	11.65
14	Oxygen	wt. %	11	–	11.44
15	Ash content	% (m/m)	Max. 0.02	Max. 0.02	~0
16	Oxidation stability	h	Min. 3	Min. 8	2.69
17	Linolenic acid methyl ester	% (m/m)	–	Max. 12.0	0
18	Polyunsaturated (> = 4 Double bonds) methyl ester	% (m/m)	–	Max. 1.00	0
19	Saponification number	mg KOH/g	Max. 370	–	199.98
20	Color	–	–	–	Greenish yellow
21	pH	–	–	–	7
22	Odour	–	–	–	Agreeable
23	Physical state at room temperature	–	–	–	Liquid

[a]mg/kg.
[b] at 20 °C.

- The theoretical percentage yield of *Styrax officinalis* L. seed oil biodiesel was found to be at 89.23% via Taguchi approach under the following reaction conditions: NaOH concentration of 0.6 wt%, methanol to oil molar ratio of 6:1, time of the reaction of 60 min, and temperature of the reaction of 60 °C.
- The experimentally determined biodiesel yield of *Styrax officinalis* L. seed oil was 88.77% at the same transesterification reaction conditions which were close to the theoretical yield as given above.
- According to the ANOVA results, the catalyst concentration and methanol to oil molar ratio have been identified as the most significant factors with the contributing factors of 78.07% and 20.32%, respectively.
- The physical and chemical properties of the *Styrax officinalis* L. seed oil biodiesel were analyzed and except some of the results, the others have met the requirements of the biodiesel standards.

Hence *Styrax officinalis* L. seed oil biodiesel could be evaluated as a novel and potential candidate to the petroleum-based diesel fuel. Moreover, it could solve the global apprehensions of the energy crisis and environmental impairment. In the light of this research, (i) other important physicochemical characteristics (ester content, free glycerol, total glycerol, methanol content, etc.) of the produced methyl ester, (ii) kinetic and thermodynamic studies on the biodiesel production from the *Styrax officinalis* L. oil and (iii) testing of *Styrax officinalis* L. seed oil biodiesel and its blends with diesel fuel in the compression ignition engines in order to obtain the engine performance, exhaust emissions, and combustion characteristics can be investigated for the future studies. They will also become a preliminary study.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Competing interests

The authors declare no competing financial interest.

Disclosure statement

No potential conflict of interest was reported by the authors.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fuel.2020.117025.

Appendix B

1. $DU = (monounsaturatedC_n{:}\ 1,\ wt.\ \%) + 2(polyunsaturatedC_n{:}\ 2,\ wt.\ \%) + 3(polyunsaturatedC_n{:}\ 3,\ wt.\ \%)$
   $DU = (21.05 + 0.21 + 0.39) + 2 * (70.56) DU = 162.77$
2. $SN = \sum \left(\frac{560xA_i}{MW_i}\right) SN = 560 * \left[\left(\frac{0.05}{200.324}\right) + \left(\frac{0.03}{228.378}\right) + \left(\frac{4.78}{256.432}\right) + \left(\frac{2.72}{284.484}\right) + \left(\frac{0.20}{340.592}\right) + \left(\frac{21.05}{282.468}\right) + \left(\frac{0.21}{310.522}\right) + \left(\frac{0.39}{338.576}\right) + \left(\frac{70.56}{280.452}\right)\right] SN = 199.984 mgKOH/goil$
3. $IV = \sum \frac{254xDxA_i}{MW_i} IV = 254 * \left[\left(\frac{1 * 21.05}{282.468}\right) + \left(\frac{1 * 0.21}{310.522}\right) + \left(\frac{1 * 0.39}{338.576}\right) + \left(\frac{2 * 70.56}{280.452}\right)\right] IV = 147.203 giodine/100g$
4. $CN = 46.3 + \frac{5458}{SN} - 0.225 IVCN = 46.3 + \frac{5458}{199.984} - 0.225 * 147.203 CN = 40.472$
5. $OS = -0.0518(DU) + 11.121 OS = -0.0518 * 162.77 + 11.121 OS = 2.690h$
6. $\ln(\eta_i) = -12.503 + [2.496 \times \ln(MW_i)] - (0.178 \times N_s) \ln(\eta_i) = \{-12.503 + [2.496 \times \ln(200.324)] - (0.178 \times 0)\} * \frac{0.05}{100} + \{-12.503 + [2.496 \times \ln(228.378)] - (0.178 \times 0)\} * \frac{0.03}{100} + \{-12.503 + [2.496 \times \ln(256.432)] - (0.178 \times 0)\} * \frac{4.78}{100} + \{-12.503 + [2.496 \times \ln(284.484)] - (0.178 \times 0)\} * \frac{2.72}{100} + \{-12.503 + [2.496 \times \ln(340.592)] - (0.178 \times 0)\} * \frac{0.20}{100} + \{-12.503 + [2.496 \times \ln(282.468)] - (0.178 \times 1)\} * \frac{21.05}{100} + \{-12.503 + [2.496 \times \ln(310.522)] - (0.178 \times 1)\} * \frac{0.21}{100} + \{-12.503 + [2.496 \times \ln(338.576)] - (0.178 \times 0)\} * \frac{0.39}{100} + \{-12.503 + [2.496 \times \ln(280.452)] - (0.178 \times 0)\} * \frac{70.56}{100} \ln(\eta_i) = 1.272386801 \eta_i = 3.569 mm^2/s$
7. $HHV = 49.43 - [0.041(SN) + 0.015(IV)] HHV = 49.43 - [0.041 * (199.984) + 0.015 * (147.203)] HHV = 39.023 MJ/kg$

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