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Biodiesel Production Using Vegetable Oil and Animal Fat Mixtures

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Abstract: In this study, the biodiesel samples were produced by using the mixtures of vegetable oils (sunflower and corn oils) and beef tallow. In the experiments, the mixtures of vegetable oils and beef tallow at different ratios were trans esterified in a batch-wise system with methanol by using sodium hydroxide as catalyst. The effects of temperature, time, amount of catalyst and vegetable oil/beef tallow ratio on biodiesel production were studied. The experiments were carried out by using amount of catalyst ranging from 0.125 to 1 wt% of feedstocks by keeping the molar ratio of 1/6 (oil to methanol) at temperatures between 40-70°C for different times ranging from 25 to 80 min. The biodiesel from the mixture of vegetable oils and beef tallow were obtained using the blends containing 0, 5, 10 and 20% of beef tallow by volume. All experiments were conducted at a fixed mixing speed of 600 rpm. The biodiesel conversion increased sharply until 0.75 wt% catalyst amount and slightly between 0.75wt% and 1 wt% with increasing reaction time for all feed stocks. Biodiesel conversions increased with increasing temperature from 40 to 60°C, but there was no significant increase after this temperature. The degree of conversion decreased as the beef tallow content increased in the mixture. The values of density and kinematic viscosity of biodiesel samples increased with an increase of beef tallow amount in vegetable oils. On the other hand, the heat values of biodiesel were similar to sunflower oil and beef tallow from which they were produced.

Key words: Biodiesel, transesterification, vegetable oil, animal fat.

Bitkisel ve Hayvansal Yağ Karışımlarından Biyodizel Üretimi

Öz: Bu çalışmada bitkisel (ayçiçek ve mısır özü) ve sığır iç yağı karışımları kullanılarak kesikli sistemde biyodizel üretimi araştırılmıştır. Deneysel çalışmalar farklı oranlarda hazırlanan bitkisel ve hayvansal yağ karışımlarından metanol kullanılarak sodyum hidroksit katalizörlüğünde gerçekleştirilmiştir. Çalışmada biyodizel üretimine katalizör miktarı, reaksiyon süresi, sıcaklık ve bitkisel/ hayvansal yağ oranı gibi değişkenlerin etkisi araştırılmıştır. Deneylerde kullanılan yağ/metanol oranı 1/6 (mol) olarak sabit tutulmuştur. Katalizör miktarı ise 0,125 ile 1% (ağırlıkça) arasında değişmektedir. Deneyler, 25-80 dakika arasında değişen farklı zamanlarda 40-70 °C arasındaki sıcaklıklarda gerçekleştirilmiştir. Sığır iç yağı bitkisel yağlarla hacimsel olarak 0, 5, 10 ve %20 oranlarında karıştırılarak deneysel çalışmalar yürütülmüştür. Tüm deneyler, 600 rpm'lik sabit bir karıştırma hızında gerçekleştirilmiştir. Bütün karışımlar için biyodizel oluşumu artan reaksiyon süresi ile birlikte katalizör miktarı arttıkça artmaktadır (%0,75' e kadar hızlı ve %0,75 ile %1 arasında yavaş). Biyodizel oluşumu sıcaklık 40 °C'den 60 °C'ye yükseldiğinde artmış, ancak bu sıcaklıktan sonra fazla değişmemiştir. Biyodizel oluşumu bitkisel yağ içerisinde sığır yağı artışıyla azalmaktadır. Biyodizel örneklerinin yoğunluk ve kinematik viskozite değerleri bitkisel yağlardaki sığır yağı içeriğinin artmasıyla artmaktadır. Bununla birlikte, biyodizelin ısı değerleri üretildikleri ayçiçek yağı ve sığır yağına benzerlik göstermektedir.

Anahtar kelimeler: Biyodizel, transesterifikasyon, bitkisel yağ, hayvansal yağ.

1. Introduction

The consuming of fuel in the world, especially in developing countries, has been increasing with an alarming rate. Studies have been concentrated on the searching of alternative energy sources to overcome this crisis. Biomass and biological sources have been used as alternative sources for clean and renewable energy production. Biodiesel and renewable diesel are the fuels gained much attraction among the various biomass-based fuels [1–3]. Although both fuels are biomass-based, they are different type of fuels.

Biodiesel consists of mono-alkyl esters obtained from different sources such as vegetable oils, animal fats, greases, wastes of oil and fats or from the mixture of different feed stocks. It is generally used as a fuel in engines and heating systems. Biodiesel is a nontoxic biodegradable biofuel and it is environmental profitable [4]. Biodiesel is produced from oils, fats or the mixtures of them with an alcohol by using suitable catalysis via transesterification process [1,5–10]. Although the relatively low cost of biodiesel producing process, the high cost of its raw materials is a major holdback for its marketing in wide range; the biodiesel took out from biomass is commonly more

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expensive than diesel gained from petroleum about 10 to 50% [11]. Fortunately, waste frying oils (WFOs) are remained less price feed stocks; as making biodiesel producing more supportable with compare to the production of fossil fuel-based diesel [12,13].

In order to increase the market value of biodiesel, it is necessary to make its production feasible. The use of less valuable raw materials such as solid fats, soap stocks and used frying oils can reduce the cost of biodiesel. However, it is not economically feasible to directly apply the transesterification process to these raw materials due to their high free fatty acid (FFA) content. Because the cold flow properties of biodiesel obtained from such sources, especially animal fats, are unacceptably poor due to their high saturation level [14,15]. Using mixtures obtained by adding animal fats to vegetable oils in certain proportions is an opportunity to produce biodiesel. Altun et al. [16], in their study investigating the physical properties of biodiesel obtained from canola and beef oil mixtures, observed that the kinematic viscosity of biodiesel increased with the increasing of beef fat in the mixture, but its density did not change much. Taravus et al. [17] reported that the physical properties of biodiesel produced from sunflower oil and beef fat mixtures improved significantly as the amount of beef fat in the feedstocks mixture decreased. Similarly, Dawi et al. [18] showed that many physical properties of biodiesel obtained from sunflower oil and beef fat mixtures changed significantly as the amount of beef fat in the mixture increased and these changes were within the standard range of biodiesel for 40% and 60% ratios (beef fat/sunflower oil). Despite several studies on biodiesel from vegetable oil and animal fat mixtures, experimental data on biodiesel production from such mixtures are still insufficient. Therefore, investigation of biodiesel production from vegetable oil and animal fat mixtures may be useful for a better understanding of biodiesel production in terms of utilization of waste animal fats.

The main purpose of this study is to investigate the production of biodiesel by using mixtures of vegetable oils (sunflower and corn oils) with beef tallow, and determine the obtained biodiesel physical properties such as density, kinematic viscosity, acid value, FFA, water content and heat value.

2. Materials and Methods

Sunflower and corn oils were used as vegetable oils while beef tallow was used as animal fat. The refined and winterized food-grade sunflower and corn vegetable oils were collected from local shops. Beef tallow was obtained from a local butcher and melted at 65 °C and filtrated before used in the experiments. Anhydrous grade (99.95 %) methanol (MeOH) was used for methanolysis while grade sodium hydroxide (NaOH) was used as catalysis. AR grade isopropyl alcohol (IPA) were toluene and were used as reagents for FFA measurements.

The transesterification reaction of the mixture of vegetable oils and animal fat was carried out in a batch system in which a 1000 mL three-necked glass reactor was used. A thermometer was placed in the first neck, while the second was equipped with a reflux condenser. The third neck was used as inlet for the reactants. A magnetic stirrer with a hot plate at a constant speed was used to heat the mixture in the reactor.

The parameters such as reaction time, temperature and the ratio of oil to catalyst were studied. In a typical run, first the total 200 mL mixture of oil and fat is charged to reactor. In order to obtain sodium methoxide, a certain amount of catalyst (NaOH) based on weight percent of blend is mixed with MeOH and heated to dissolve at 60°C. The molar ratio of oil to MeOH was1/6 in all experiments. The sodium methoxide is then charged to the reactor containing the oil/fat mixture. The mixture was then stirred continuously at desired temperature maintained by controlling the electrical heating till a required time. After the completion of the transesterification reaction, the mixture in the reactor was kept in a separating funnel for at least 24 hours to separate the fatty acid methyl esters and glycerol phases. After the separation of the phases from each other, the ester phase was washed 3-4 times with warm distilled water to remove excess unreacted methyl alcohol and glycerin. A few drops of 0.1% H_2SO_4 were added to remove trace amounts of catalyst in the ester phase and the washing process was carried out again. The washing process was repeated until a clear water layer with neutral pH was obtained. A rotary evaporator was used to remove excess methanol that might have been carried over during the washing process. The amount of catalyst used varied between 0.125 and 1% of the mixture by weight. The temperatures studied were varied from 40 to 70°C and the times were between 25 and 80 minutes. All experiments were carried out at a fixed mixing speed of 600 rpm. All experiments were conducted at least duplicate for the reproducibility of the process.

The chemical compositions of raw materials and biodiesels were performed by using a GC analyzer (GC- 6C 2010 plus model by Shimadzu Inc., Kyoto, Japan). The capillary column was 0.25 mm in diameter and 100 m long. Each fatty acid methyl ester determination was run in triplicate and average values are used. Molecular weights of sunflower and corn oils, beef tallow, and biodiesel samples were calculated using Equation (1):

$$M_w = \sum M_{Wi} x_i \quad (1)$$

where, M_w is the molecular weight of oil, fat or biodiesel, M_{wi} is the molecular weight of individual methyl esters and x_i is the mole percent of fatty acid methyl ester.

The biodiesel physical properties such as density, kinematic viscosity, acid value, FFA, water content and heat value were determined using appropriate standard test methods. The density of biodiesel samples was determined using a 25 mL density bottle. The density bottle completely filled with sample was weighed on a precision electronic analytical balance and the density was calculated from the weight and volume of the bottle at 15°C. The kinematic viscosities of the samples were measured by Canon Fenske Routine (PSL ASTM-IP 75) viscometer. The test sample was heated to 40 °C in the viscometer. The time elapsed for a certain volume of sample poured from cab of the viscometer was recorded. This value was the converted to kinematic viscosity using the viscosity table. The acid number and FFA content of sunflower oil, corn oil, beef tallow and biodiesel were measured by following ASTM D664. 5 grams of sample was dissolved in 125 mL of 50% toluene and 50% IPA (v/v) solution. The prepared solution was titrated with 0.1 M KOH solution with 1% alcohol as phenolphthalein indicator. The water content of the feedstocks and biodiesel produced was determined using a Mettler Toledo Karl Fischer DL 31 Titrator. The calorific value of the samples was measured by using an oxygen bomb calorimeter (11350 automatic adiabatic model, Julian Peters Co., Moline, IL, USA). All the procedures for measurements of physical properties were repeated three times for process reproducibility.

3. Results and Discussion

3.1. The Characteristics of Feedstocks

The chemical compositions and some properties of vegetable oils and beef tallow are shown in Table 1 and Table 2, respectively. As seen in Table 1, the beef tallow methyl ester's saturated fatty acid amount is 42.57% while saturation degree is 9.79% for sunflower oil methyl ester 14.55% for corn oil. Due to the effect of the degree of saturation in oils and fats on fuel properties, the saturation value in feedstocks can be important when the produced biodiesel is used as fuel in diesel engines.

It can be seen from Table 2 the oils and fat used in the study have typical physical properties as most oils and fats indicate. The data of Table 2 show that sunflower and corn oil samples contained lower percentage FFA of 0.226% and 0.197%, respectively. However, the beef tallow sample contained slightly higher percentage FFA of 0.533%. Feedstocks containing high FFA cause soap formation as side reaction in biodiesel production. Soap formation can cause difficulty in the separation of biodiesel and glycerol phases and thus affect biodiesel conversion and purity.

3.2. Effects Amount of Catalyst and Time on Biodiesel Production

The amount of catalyst and time for the transesterification process are important factors that could affect the biodiesel conversion [19]. In order to examine the effect of catalyst amount on the biodiesel production, experiments were performed for pure sunflower oil and corn oil using 60 °C reaction temperature and 60 min reaction time as a function of different percentage of catalyst. Figure 1 shows the biodiesel conversion as a function of catalyst percentage. It can be seen from Figure 1, the biodiesel conversion increases as the catalyst percentage increases. The Figure also shows that about 97% biodiesel conversion for sunflower oil and 96 % for corn oil were observed when 1 wt% NaOH was used.

The amount of NaOH was in the range of 0.25–1.25 wt% of vegetable oil-tallow mixtures in order to investigate effects of time and catalyst amount. Figure 2 shows the variation of the biodiesel conversion with the reaction time at different catalyst concentration using 10% tallow in sunflower oil at the constant temperature of 60 °C. As can be seen from Figure 2, the biodiesel conversion increased with increasing time for all the catalyst amount used. Similar results were obtained when using corn oil as a feedstock (Figure 3). The optimal conversions for both oils biodiesel and their mixtures with beef tallow were obtained at 60 min reaction time for each catalyst amount. It can be noticed that for reaction times smaller than 60 min the conversions were lower. The results can be explained by the fact that reaction time lower than 60 min is not enough for methanol to complete the transesterification of all the triglycerides contained in oil. When the reaction time exceeded 60 min, there is small increase in the conversions. On the other hand, when increasing the catalyst amount in the reaction mixture, the biodiesel conversion increased sharply until 0.75 wt% catalyst amount and slightly between 0.75 wt% and 1 wt% with increasing reaction time for both oils used (Figures 2 and 3). From these results, it can be said that the

Table 1. Fatty acid methyl ester compositions of oils and fat used in this study.

Ester	Sunflower oil	Corn oil	Beef tallow
Methyl myristate	0	0	3.72
Methyl palmitate	6.54	11.97	23.85
Methyl palmitoleate	0	0	1.56
Methyl stearate	3.25	2.58	14.75
Methyl oleate	27.84	25.12	47.32
Methyl linoleate	60.91	58.72	6.31
Methy linolenate	0.2	0.65	0.2
Methy arachnidate	0	0	0.25
Others	1.26	0.98	2.04
Saturated	9.79	14.55	42.57
Unsaturated	88.75	84.52	55.39

Table 2. Some properties of oils and fat used in this study.

Property	Units	Sunflower oil	Corn oil	Beef tallow
Molecular weight	g/mole	875	869	855
Density (at 15 °C)	g/cm^3	0.92	0.91	0.89
Kinematic viscosity (at 40 °C)	mm^2/s	32.5	33.1	170.1
Acid value	mg KOH/g	0.449	0.393	1.061
% FFA	%	0.226	0.197	0.533
Water content	%	< 0.05	< 0.05	< 0.05
Calorific value	MJ/kg	39.6	39.5	40.5

conversion of biodiesel is strongly dependent on alkali catalyst amount and the optimum amount of NaOH of 1 wt% could be acceptable value for both vegetable oils used. Time and amount of catalyst found in this study were close to the literature values obtained for both oil biodiesel and similar vegetable oil diesel [20–23].

3.3. Effect of Temperature on Biodiesel Production

The effect of temperature on biodiesel production was studied with the mixture of 10% beef tallow in vegetable oils by using catalyst amount of 1 wt%. Figures 4 and 5 show the effect of biodiesel conversion on the reaction temperature in the presence of 1 wt% NaOH amount for sunflower oil-beef tallow mixture and corn oil-beef tallow mixture, respectively. As seen from Figures 4 and 5, increasing the temperatures from 40°C to 60°C increased the biodiesel conversion. However, when temperature increased 70ºC, the methyl esters conversion decreased. This is because high temperature enhances soap formation as side reaction. At 60 min biodiesel conversion of both vegetable oils used were nearly highest values for all temperatures studied. The results are in agreement with those that found in the literature [20,24–27]. It can be concluded that temperature is an important parameters affecting the reaction rate and biodiesel conversion and it can be seen that 60ºC is the optimal temperature for both vegetables studied in this study.

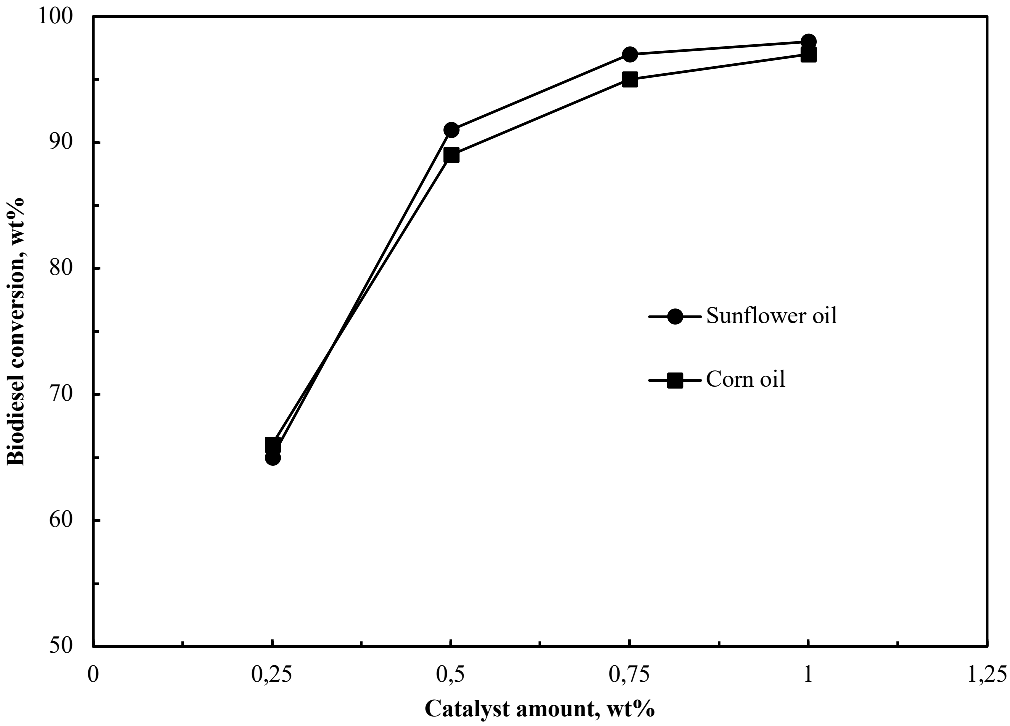

Figure 1. Effect of catalyst amount on biodiesel conversion obtained from pure vegetable oils (60 °C and 60 min).

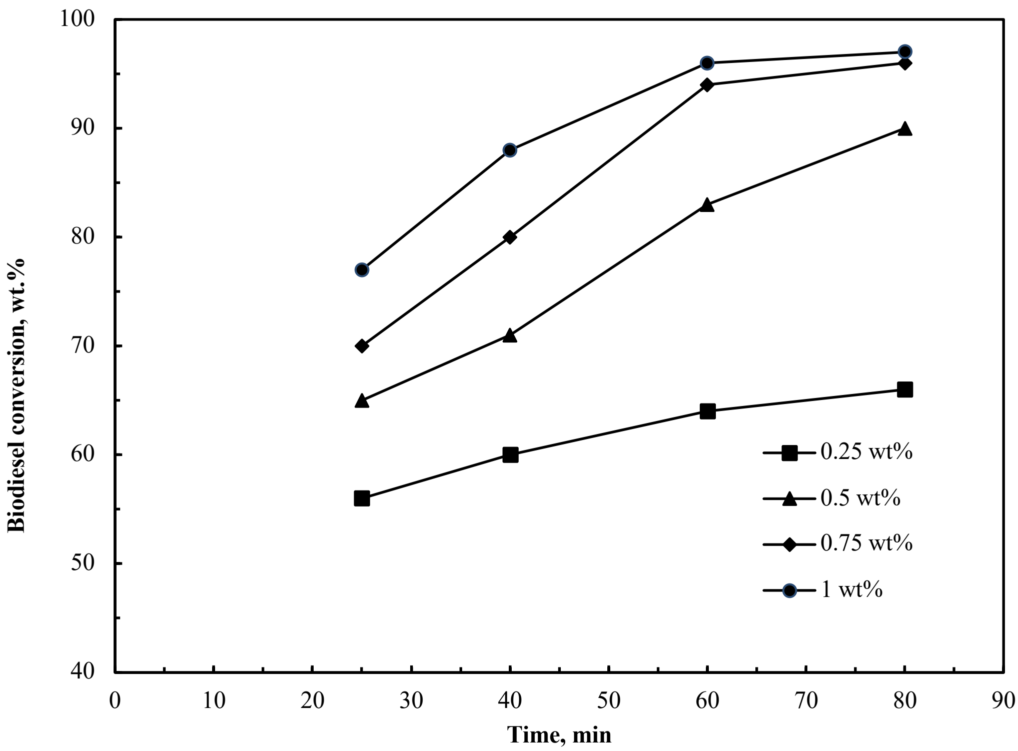

Figure 2. Effect of time on sunflower oil biodiesel conversion at different catalyst amount (60 °C and 10 % beef tallow).

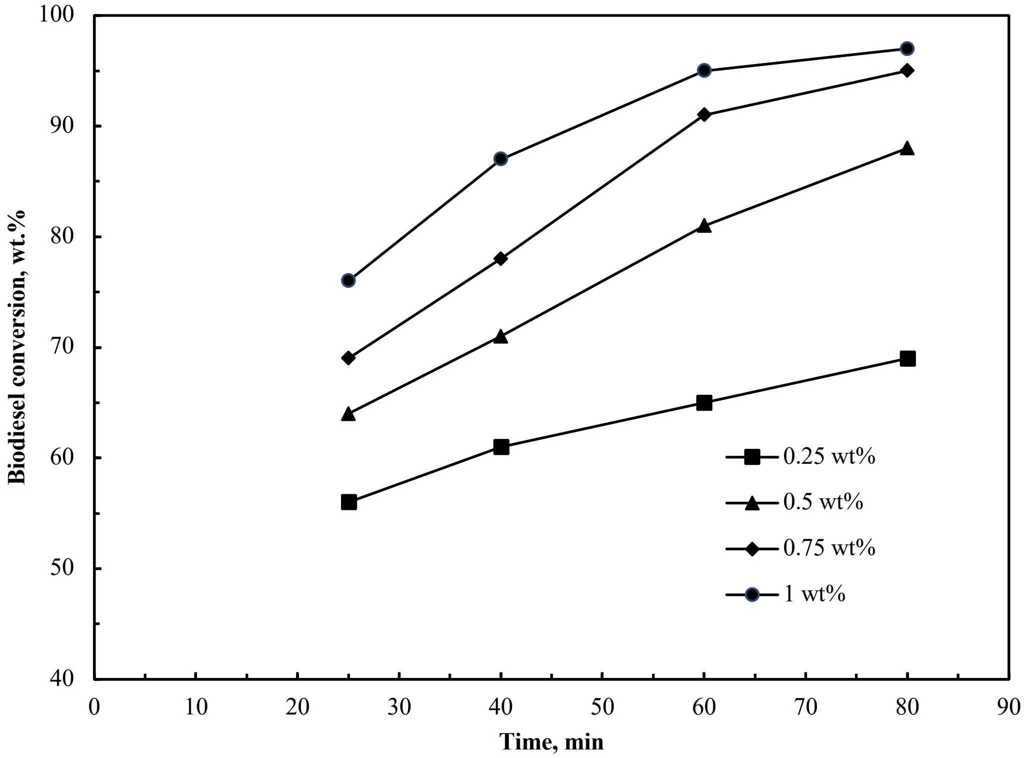

Figure 3. Effect of time on corn oil biodiesel conversion at different catalyst amounts (60 °C and 10 % beef tallow).

3.4. Effect of Beef Tallow Amount on Composition and Physiochemical Properties of Biodiesel

The transesterification of sunflower and corn oils, and their mixtures with beef tallow were carried out using the blends containing 0, 5, 10 and 20% of beef tallow in volume basis to examine effects of beef tallow amount in the mixture on the conversion and properties of biodiesel. The experimental conditions were set up as reaction time of 60 min and 1wt%. Table 3 shows the fatty acid methyl ester compositions, saturation degrees and conversions of biodiesel produced using vegetable oils-beef tallow mixtures. The amounts of methyl esters of oleic acid, palmitic acid, and stearic acid increase with the animal fat content, whereas methyl esters of linoleic acid and linolenic acid decrease as the beef tallow content increases in the sunflower oil (Table 3). Similar results were also seen in Table 3 for corn oil. As can be noted that content and saturation degree of the fatty acid methyl esters in the biodiesel corresponded to the weighted average of its component content in the mixtures. The biodiesel conversion decreases as the beef tallow content increases in the vegetable oils used. This can be attributed to the high FFA content of the beef tallow used (see Table 2). Because the oil samples with high FFA content consume more alkali catalyst to neutralize the FFA in the reaction mixture [14]. The oil samples having high FFA enhance soap formation as side reaction which could cause difficulty in the phase separation of the biodiesel, and affect the yield and the purity of the alkyl esters produced. At the optimum operating conditions, the conversions of biodiesel (methyl esters) from all the mixture of vegetable oil and beef tallow studied varied from 91% to 97.1%. On the other hand, it was observed that the methyl esters conversions obtained for the mixture of sunflower oil and beef tallow are higher than those obtained for the mixtures of corn oil and tallow. Previous studies on the production of biodiesel from the common vegetable oils such as sunflower oil [19,24,28], corn oil [2,29], soybean [2,19,30], canola oil [2,11] and rapeseed [28] gave biodiesel conversions over 90 %. On the other hand, nearly 90% biodiesel conversions were obtained for pure beef tallow [22,31] and for the mixture of vegetable oils and beef tallow [17,18,20]. Considering the studies given above, the conversions obtained in this study are within the range of them.

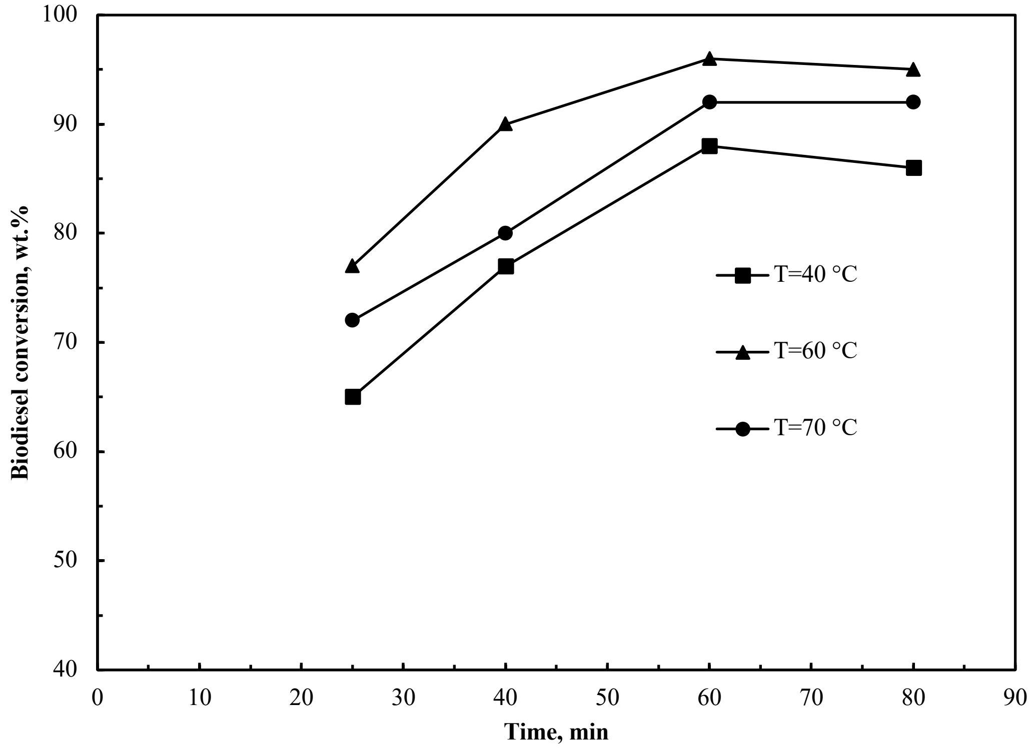

Figure 4. The effect of temperature on conversion of biodiesel obtained from sunflower oil-beef tallow mixture (1 wt% catalyst amount and 10 %beef tallow).

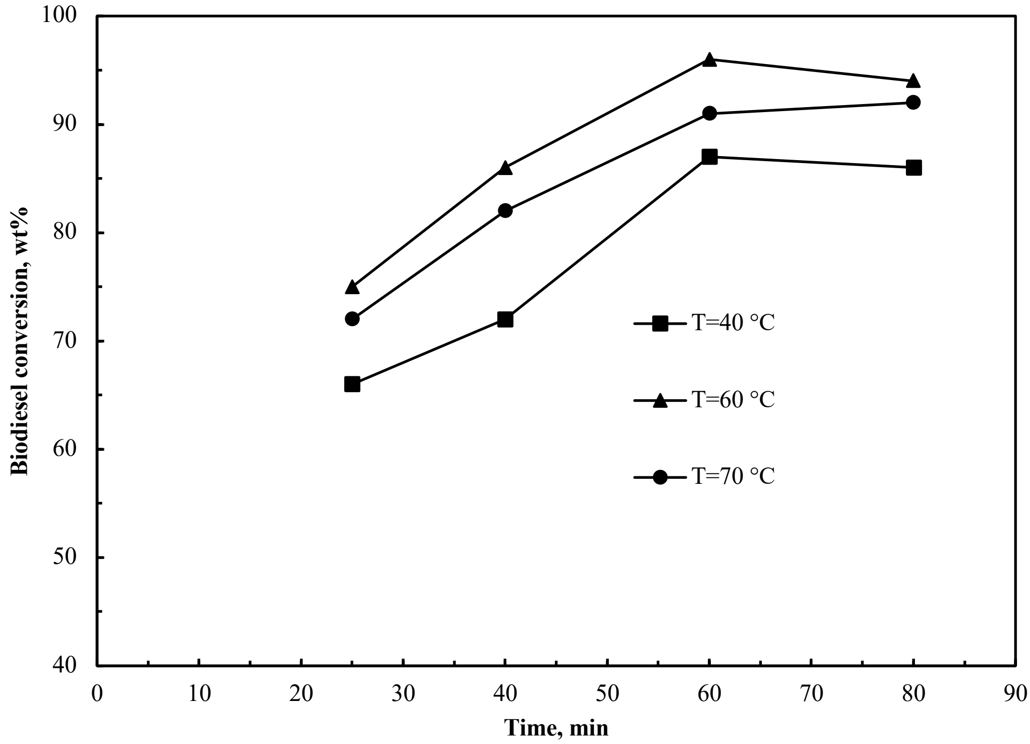

Figure 5. The effect of temperature on conversion of biodiesel obtained from corn oil-beef tallow mixture (1 wt% catalyst amount and 10% beef tallow).

Table 4 summarizes the physical properties of biodiesel produced using the mixtures of sunflower-beef tallow and corn oil-beef tallow. The density has significant effect on the engine combustion system. Because the lower density of fuel breaks up the fuel injected into the combustion. It is known that the density of biodiesel is considerably lower than feedstocks which from it is produced. The density of biodiesel samples produced decreased smoothly with the increase of amount of beef tallow in vegetable oils (Table 4). It was reported that the densities of vegetable oils did not change too much during transesterification process since the densities of methanol and oils are similar to the density of the produced biodiesel and for the blends of vegetable oils with animal fats, the density of methyl ester was reported in the range of 870-890 kg/m^3 [32]. The viscosity is another important property of biodiesel and should be lower than that of its source. The values of viscosity of the biodiesel samples increased with the increasing of beef tallow amount in the vegetable oils. However, the value of kinematic viscosity was slightly high for the mixtures using 20% beef tallow for both vegetable oils to fulfill the standard limits of the standards. This is because of the pure beef tallow as feedstock which have the high viscosity [33,34]. Similar results were reported by Taravus et al. [17] for the mixture of sunflower oil and beef tallow and by Adin et al. [35] for the mixture of canola oil and beef tallow. However, the kinematic viscosity values of the obtained biodiesel for 0, 5 and 10% beef tallow are between the standard limit values [33]. As seen from Table 4, the acid number and acid content decrease with increasing beef tallow content in vegetable oil. The acid content of biodiesel fuel can be come out from FFAs of feedstocks or excess acid that is not used in the biodiesel production process. The acid values and the values of acid contents for all biodiesel samples produced in this study met the standards [33]. The heat values of the biodiesel from vegetable oils-beef tallow mixtures are summarized in Table 4. It is seen from Table 4 that the heat value decreases with increasing beef tallow content in the vegetable oils. On the other hand, the values heat of biodiesel was similar to vegetable oils and beef tallow from which they were produced (see Table 2).

As mentioned above, when comparing both biodiesel conversion and physical properties with literature values, it is seen that biodiesel obtained by blending vegetable oils with low proportions of beef tallow is a suitable fuel. It is reported that the raw material (vegetable oil or animal fat) in biodiesel production constitutes approximately 70–95% of the total cost [9,11,18]. Therefore, it may be economically feasible to obtain high-yield biodiesel by adding animal fats to vegetable oils.

Table 3. Fatty acid methyl ester compositions of biodiesel produced by using mixtures of vegetable oils and beef tallow.

Ester	Sunflower oil				Corn oil			
	Beef tallow (%)				Beef tallow (%)			
	0	5	10	20	0	5	10	20
Methyl myristate	0	0.17	0.39	0.72	0	0.14	0.36	0.73
Methyl palmitate	6.54	7.38	8.17	9.98	11.97	11.95	13.31	14.28
Methyl palmitoleate	0	0.08	0.16	0.3	0	0.079	0.14	0.31
Methyl stearate	3.25	4.05	4.85	5.61	2.58	3.22	3.41	3.52
Methyl oleate	27.84	27.81	29.15	31.5	25.12	26.21	27.52	29.34
Methyl linoleate	60.91	59.18	56.02	50.51	58.72	56.61	53.28	49.94
Methy linolenate	0.2	0.23	0.22	0.21	0.65	0.61	0.58	0.56
Methy arachnidate	0	0.013	0.025	0.05	0	0.013	0.025	0.03
Others	1.26	1.07	1.01	1.12	0.98	1.168	1.375	1.29
Saturated	9.79	11.61	13.43	16.36	14.55	15.32	17.11	18.56
Unsaturated	88.75	87.07	85.32	82.28	87.45	83.51	81.52	80.15
Methyl esters conversion (wt%)	0.971	0.96	0.95	0.93	0.96	0.95	0.93	0.91

Table 4. Some properties of biodiesel produced from mixtures of vegetable oils and beef tallow.

Property	Sunflower oil				Corn oil			
	Beef tallow (%)				Beef tallow (%)			
	0	5	10	20	0	5	10	20
Density at 15 °C, g/cm^3	0.877	0.877	0.874	0.871	0.878	0.877	0.876	0.875
Kinematic viscosity at 40 °C, mm^2/s	4.89	4.92	4.98	5.25	4.53	4.58	4.67	5.32
Acid value, mg KOH/g oil	0.281	0.365	0.224	0.282	0.449	0.331	0.387	0.378
Acid content, %	0.141	0.183	0.113	0.145	0.226	0.166	0.195	0.190
Heat value, MJ/kg	40.18	39.29	38.14	39.86	38.66	38.29	37.14	37.86
Water content, %	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05

4. Conclusions

The mixtures of vegetable oils and animal fat biodiesels were successfully produced by means of chemical transesterification using MeOH and sodium NaOH. The biodiesel from the mixtures of vegetable oil (sunflower and corn oils) with beef tallow were obtained using the blends containing 0, 5, 10 and 20% of beef tallow in volume basis. It can be concluded from this study that the production of biodiesel from the mixture of vegetable oil sources and beef tallow gave considerable high biodiesel conversions and acceptable physical properties. Although the yield of biodiesel obtained from edible vegetable oils is high, the threat it poses to human food resources makes biodiesel production from these sources unsustainable. For this reason, the addition of unused animal fats to vegetable oils in appropriate amounts can be a reliable and affordable raw material for biodiesel production. It is also necessary to take advantage of these kind of sources to reduce the raw material cost to produce biodiesel. In the view of the results of present study, it is recommended that biodiesel fuel without lowering biodiesel quality can be produced by using different kind of mixture of non-edible oils and relatively less expensive animal fats such as chicken fat, lard and fish oil.

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