



Characteristics of direct transesterification using ultrasound on oil extracted from spent coffee grounds

Yeong Su Kim¹, Duk Gam Woo^{2†}, Tae Han Kim^{1*}

¹Department of Bio-industrial Machinery Engineering, Kyungpook National University, Daegu, 41566, Korea

²Research Faculty of Agriculture, Hokkaido University, Sapporo, 060 - 8589, Japan

ABSTRACT

Spent coffee grounds (SCG), the residue after brewing coffee beverage, is a promising biodiesel feedstock due to its high oil contents (15–20%). However, SCG should be pretreated to reduce the high free fatty acid content, which hampers transesterification reaction. To overcome this, we explored a direct transesterification reaction of SCG using ultrasound irradiation and identified the optimal sonication parameters. A high fatty acid methyl ester (FAME) content, up to 97.2%, could be achieved with ultrasound amplitude of 99.2 μm , irradiation time of 10 min, and methanol to oil ratio of 7:1 in the presence of potassium hydroxide concentration of 1.25 wt.%. In addition, we demonstrated that ultrasound irradiation is an efficient method to produce biodiesel from untreated SCG in a short time with less energy than the conventional mechanical stirring method. The physical and chemical properties of the SCG biodiesel met the requirements for an alternative fuel to the current commercial biodiesel.

Keywords: Biodiesel, Energy consumption, Pretreatment, Spent coffee grounds, Transesterification, Ultrasound

1. Introduction

Biodiesel (BD) produced mainly from vegetable oils is the most promising and practical alternative energy for transportation because it can be produced, distributed, and stored using the current power units and infrastructure. The use of vegetable oil is limited due to demand. Also, potential feed shortage and increase in feed price further limits its usage [1]. In this regard, research is ongoing for BD that can be produced from non-feed and industrial waste materials. Spent coffee grounds (SCG) are a promising and practical BD feedstock [2].

Coffee is one of the most popular beverages worldwide. In 2017, annual consumption amounted to 97 million tons, with trading volume second to that of petroleum [3, 4]. Noting that 2 kg of SCG is produced for each 1 kg of coffee, Nestle, a global coffee manufacturer has been exploring various approaches to using SCG for the production of compost, solid fuel, and BD [5, 6]. The oil content of SCG is in the range of 15–20%. Efficient conversion of this high oil content into BD would reduce the costs of SCGs disposal and BD production, making this promising alternative energy more economically feasible [7].

The oil extracted from SCG contains 2.01–5.02% free fatty acids

(FFA), which facilitates saponification and water production during the transesterification process, consequently reducing BD yield [6]. In general, efficient alkali-catalyzed transesterification reaction requires FFA content less than 1.0% [8]. According to previous study, the FFA content needs to be less than 0.5% for alkaline transesterification to occur [9]. For transesterification of low-grade oils such as SCG, waste cooking oil, and animal fat, the feedstock should be pretreated using an acid catalyst to remove FFA [10]. The pretreatment process is usually carried out at high pressure and temperature in the presence of an acid catalyst, increasing energy consumption and adding wastewater treatment cost to the BD price [11]. The cost of the pretreatment process performed to remove FFAs from low-grade raw materials increases the BD production cost. According to Y. Zhang et al. [12], although the higher price of virgin oils is 2.5 times higher than that of waste cooking oils, the break-even price of waste cooking oil-derived BD is approximately 3% higher than that of virgin oil-derived from BD due to the higher pretreatment cost of waste cooking oils. Therefore, a novel approach is required to reduce the cost and energy invested in the pretreatment process.

To address this issue, many recent studies have attempted to take advantage of ultrasound for SCG pretreatment [13] and activa-



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† Corresponding author

Email: dgwoo@bpe.agr.hokudai.ac.jp

Tel: +81-11-706-2539 Fax: +81-11-706-4147

ORCID: 0000-0003-0281-4342

tion of the transesterification reaction. Ultrasound irradiation is reported to facilitate mass transfer across interface during chemical reactions by inducing cavitation [14]. Ultrasound has been used for the production of BD from plant oils, including soybean oil, canola oil, corn oil, sunflower oil, coconut oil [2, 14-17], and non-food feedstock, including waste cooking oil, beef tallow, and SCG [18-20].

Hingu et al. [18] investigated the potential use of ultrasound for the transesterification of waste cooking oil and examined the influence of ultrasound power, potassium hydroxide (KOH) concentration, the molar ratio of methanol to oil, and reaction time on the FAME content of BD. The ester content could be increased up to 89.5% when the waste cooking oil was sonicated at 45°C for 40 min with ultrasound power of 200 W, methanol to oil molar ratio of 6:1, and KOH concentration of 1.0 wt.%. Compared to a mechanical stirring method at an equivalent condition, the ester content was 32% higher [18].

Teixeira et al. [19] used ultrasound for the transesterification of beef tallow (24 kHz, 400 W). When the transesterification was carried out with methanol to oil molar ratio of 6:1, KOH concentration of 0.5 wt.%, and reaction time of 70 s at 45°C, the ester conversion rate was 92%, which was comparable to the 91% achieved using a mechanical stirring method. In their study, improvement in reaction time by the use of ultrasound was found: the reaction time required for equivalent conversion was reduced significantly (to < 1 h) [19].

Liu et al. [21] attempted a direct conversion of SCG, instead of using the oil extracted from SCG, to BD using an in-situ BD process. They experimented with catalyst concentration, reaction time, and temperature. The highest FAME content was 95.3% in the transesterification performed at 70°C for 12 h in the presence of 20 wt.% H₂SO₄. This FAME content is below the level specified in the EN14103 standard, and this approach suffers from a higher acid catalyst concentration and longer reaction time. The amount of acid catalyst was 20 times higher than that of the conventional acid-catalyzed transesterification (1.0–5.0 wt.% H₂SO₄) [22] and required substantially longer reaction times (> 10 h) [23].

Burton et al. [24] compared the BD conversion rates by KOH-catalyzed transesterification and enzymatic transesterification reactions. The SCG samples were pretreated with hydrochloric acid, methanol and oil mixture (40:1 mole/mole) for 2 h. For the KOH-catalyzed transesterification, the conversion rate was less than 90% when the reaction was allowed at 55°C for 1 h with 1.0 wt.% catalyst and 6:1 of methanol to oil molar ratio. For the enzymatic transesterification, the BD conversion rate was increased up to 98.5% when the reaction was allowed to proceed at 35°C for 72 h with 2.0 g enzyme. During the transesterification reaction, a mixture of methanol to oil (3:1 mole/mole) was added. After a specified time, the same mixture, but with a different molar ratio (1:3) was added. Al-Hamamre et al. [25] achieved a BD conversion of 85.5% in one-step transesterification reaction at 65°C within 4 h. The molar ratio of MeOH-KOH to FFA was 9:1. In a two-step reaction, they first pretreated SCG with 1.0 wt.% sulfuric acid and 10-20 mL methanol for 4 h. In the following transesterification reaction, the BD conversion rate of 99% was achieved at 60°C within 6 h with a 9:1 molar ratio of MeOH to FFA and 1.5 wt.% of KOH.

Rocha et al. [20] used an ultrasonic bath (40 kHz, 160 W) for

SCG pretreatment and transesterification. The highest ester content was 97% with H₂SO₄ 3.5 wt.%, methanol to oil molar ratio of 9:1, and reaction time of 60 min at 60°C. However, to date, the impact of the sonication parameters, such as ultrasound amplitude and irradiation time, on the transesterification of SCG oil have not been elucidated.

In this study, we identified the optimal ultrasound amplitude and irradiation time (reaction time) for efficient ultrasound-assisted transesterification, and various methanol to oil molar ratio and catalyst concentration were investigated to achieve the highest FAME content at a given sonication condition. In addition, the energy consumption and material costs for various BD production methods were estimated to analyze the economic feasibility of the ultrasound-assisted BD production. Furthermore, the physical and chemical properties, including cetane number, oxidation stability, and sulfur content, of the SCG BD were determined to evaluate the feasibility as an alternative to the current commercial BD.

2. Materials and Methods

2.1. Materials and Extraction Process

The SCG used in this study was collected from retail coffee stores, and dried at 105°C for 24 h before extracting the oil. Dried SCG (2 kg) was placed in an oil press machine (BDF-200M, Shinjin Grab Industrial Co., Korea) and pressurized at 600 kgf/cm² and 160°C for 30 min. The expressed oil was filtered to remove impurities using a glass fiber filter. On average, 159 g of oil was extracted from 2 kg of SCG. A transesterification reaction was carried out using 50 g of SCG oil.

2.2. Determination of Acid Value

The Acid Values (AV) of extracted SCG oil and waste cooking oil were determined and compared. AV was measured according to the method described in the Korean Standard (KS H ISO 660), and FFA content was calculated using Eq. (1). SCG was pretreated to remove FFA in the mixture of methanol (25 wt.%) and H₂SO₄ (0.5 wt.%) at 65°C and 250 rpm for 1 h. After washing with distilled water to remove residual methanol and sulfuric acid, the treated SCG was dried before transesterification.

$$\text{FFA (\%)} = \text{AV} \times 282/56.1 \times 100/1000 \quad (1)$$

2.3. Transesterification of SCG

The transesterification of SCG was performed using either mechanical stirring or ultrasound irradiation. In the mechanical method, the transesterification reaction was allowed to proceed for 1 h at 55°C and 250 rpm. The methanol to oil molar ratios and KOH concentrations were varied to identify the optimal condition (Table S1).

In ultrasound-assisted transesterification, the methanol to oil molar ratio and KOH concentration were fixed to 7:1 and 1.5 wt.%, respectively, and the ultrasound amplitude and irradiation time (reaction time) were varied same as above as summarized in Table 1. Previous studies reported that the increase in reaction temperature due to ultrasound-induced cavitation evaporates methanol, resulting

Table 1. Test Variables for Using Ultrasound to Transesterification

Contents (unit)	Variable
Ultrasound irradiation amplitude (μm)	24.8, 43.4, 62.0, 80.6, 99.2, 117.8
Ultrasound irradiation time (min)	5, 10, 20, 30, 40, 50, 60

in lower BD conversion rates at reaction temperatures higher than 55°C [26, 27]. To avoid rapid temperature increase due to continuous ultrasound irradiation, the samples were kept in a water bath at 55°C during the reaction.

To investigate the influence of the methanol to oil molar ratio and catalyst concentration on ultrasonic transesterification, methanol to oil molar ratio was varied from 3:1 to 11:1 and catalyst concentration from 1.0 to 2.0 wt.% (same for mechanical transesterification). Once the reaction was completed, BD was separated from glycerol using a separation funnel. Then, BD was washed with distilled water to remove any potential residuals and heated to 110°C to remove moisture.

The sonication was performed using an ultrasonic processor (VCX-750, Sonics & Materials Inc., USA, 750 W, 20 kHz) equipped with a solid-type horn (13-mm diameter, 139-mm length, and maximum amplitude of 124 μm). A heating stirrer (Corning PC-420D, Corning, USA) was used. Transesterification reaction was carried out using anhydrous methanol (99.8%) in the presence of KOH (85%). All transesterifications were repeated five times, and the reported results represent the averages of the five measurements.

2.4. Energy Consumption and Input Cost for Transesterification

An energy meter (HPM-100A, AD power, Rep. Korea) was used to measure the power consumption of the units used in the processes [28–30]. For each, the amount of energy consumed was measured five times, and the average values were used. The unit costs presented in Table S2 were used to determine the energy and material cost for BD production. The reaction condition that yielded the highest conversion rate was referred to in the energy and material cost analysis. The SCG cost was US\$ 0.02/kg [31], and for electric power, the industrial electricity price provided by Korea Electric Power Corporation (KEPCO) was used [32]. The unit costs of catalysts and solvents, including methanol and KOH, were calculated based on the actual purchase price. Therefore, it should be noted that the material cost estimated in this study was higher than the industrial production cost, where the material cost can be lowered by large purchase volume [12, 33, 34].

2.5. Analysis of SCG BD

The FAME content produced from SCG oil was determined following the method described in the EN 14103 standard. Gas chromatography (GC 6850, Agilent Technologies, Inc., USA) equipped with FID and HP-Innowax column (30 m \times 320 mm \times 0.25 μm) was used for the FAME analysis. The internal standard was prepared by adding 0.5 g methyl heptadecanoate (C17) into a volumetric flask containing 50 mL heptane (10 mg/mL). Helium was used as the carrier gas. The temperatures of the inlet and FID were set to 250 and 300°C, respectively. The oven temperature was maintained at 210°C for 9 min and increased to 230°C at the rate of

20°C/min and maintained for 10 min. The injection volume was 1.0 μl (80:1 split mode).

The physical and chemical properties of the SCG BD were analyzed at the Korean Petroleum Quality and Distribution Authority (K-Petro) using the test standards summarized in Table S3.

3. Results and Discussion

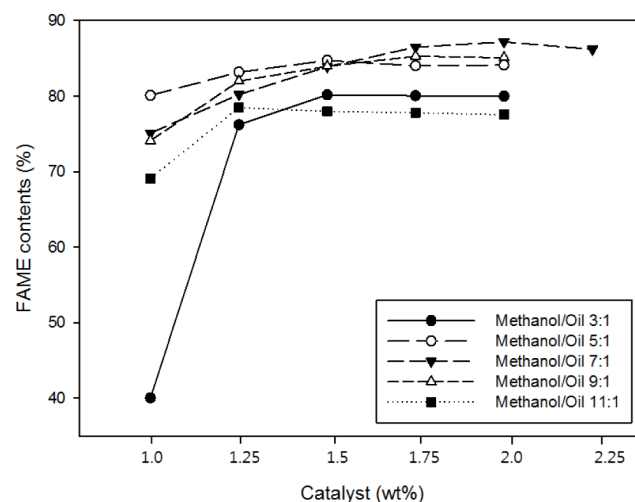
3.1. Evaluation of Acid Value

The AV of SCG oil was compared to that of waste cooking oil. The results before and after the pretreatment are summarized in Table S4. The AV of the untreated waste cooking oil were between 1.4 and 3.73 mgKOH/g (FFA 0.7–1.86%) and after the pretreatment, decreased to 0.327 mgKOH/g (FFA 0.16%).

The AV of untreated SCG oil was 6.3–8.5 mgKOH/g (FFA 3.15–4.25%), which was higher than that of the waste cooking oil (low-grade BD feedstock) by 2.4–2.5%. FFA contents higher than 1.0% hampers the separation of ester from glycerin in alkali-catalyzed transesterification reaction, lowering overall ester conversion rate [10, 35].

3.2. Transesterification Using Mechanical Stirring

Fig. 1 shows the effects of methanol to oil molar ratio and catalyst concentration used in mechanical transesterification on the FAME content. When methanol to oil molar ratio was fixed to 7:1, the FAME content increased with the catalyst concentration up to 2.0 wt.%. No further improvement was observed at a higher catalyst concentration. Similarly, in all other molar ratios, the FAME content peaked at specific catalyst concentrations. The maximum FAME content in mechanical transesterification was 87.8% with the methanol to oil molar ratio of 7:1 and 2.0 wt.% catalyst concentration. At this condition, the BD yield was 56.2%. The SCG BD produced by mechanical transesterification failed to meet the BD quality standard (FAME content \geq 96.5%). This result could be ascribed to the high FFA content of untreated SCG oil.

**Fig. 1.** Effect of catalyst concentration and methanol to oil molar ratio on the FAME content.

3.3. Transesterification Using Ultrasound Irradiation

3.3.1. Effect of ultrasound amplitude and ultrasound irradiation time

Fig. 2 shows the FAME content of BD produced by ultrasound-assisted transesterification. Regardless of the ultrasound amplitude tested, the irradiation time of 10 min was optimal for higher FAME contents. Further increase in reaction time resulted in lower FAME contents. As the FAME content increases, the hydrolysis of ester also increases and finally, dynamic equilibrium is established [17, 36]. In addition, when the reaction is allowed for an extended time, the reverse reaction can limit further transesterification, lowering the product yield and increasing soap formation [37]. The reduced reaction time by ultrasound irradiation can be associated with the ultrasonic cavitation. This provides sufficient energy to activate the transesterification of the SCG oil and increase the reactivity by improving alcohol-oil mixing [38]. With respect to ultrasound amplitude, the FAME content increased in the range of 24.8–99.2 μm . A slight decrease in the FAME content was observed at higher amplitude (117.8 μm). High ultrasound amplitudes induce the cushioning effect, which reduces the energy available for the activation of the transesterification reaction [18]. In summary, the optimal amplitude and irradiation time were 99.2 μm and 10 min, respectively, and the combination of these parameters produced the highest FAME content of 90.8%. The FAME content was significantly lower (34.2–41.6%) with the amplitude of 24.8 μm .

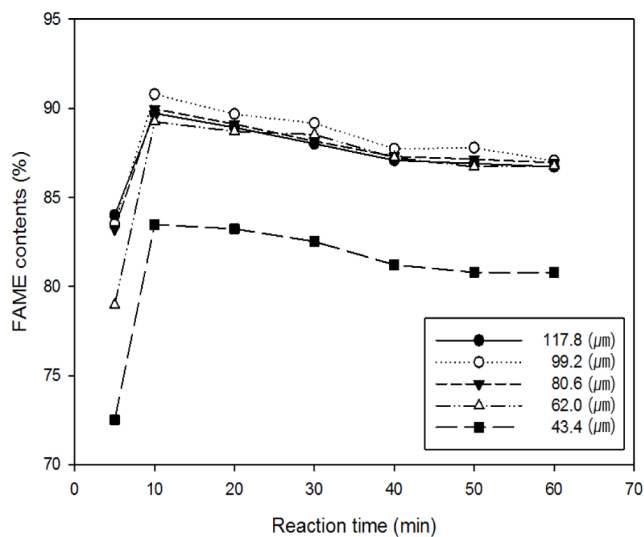


Fig. 2. Effect of ultrasound amplitude and reaction time on the FAME content with methanol to oil molar ratio of 7:1 and catalyst concentration of 1.5 wt.%.

3.3.2. Effect of methanol to oil molar ratio and catalyst concentration

Fig. 3 shows the effects of the methanol to oil molar ratio and catalyst concentration on the transesterification of SCG oil at the optimal sonication condition. The highest FAME content was 97.2% with methanol to oil molar ratio of 7:1 and catalyst concentration

of 1.25 wt.%. The BD yield at this condition was 74%. The FAME content increased as the methanol to oil molar ratio increased from 3:1 to 7:1. Further increase of methanol negatively affected the FAME content. Overload of methanol hinders proper phase separation of ester and water, and methanol can emulsify the reaction mixture, reducing the ester conversion [18, 37]. With respect to catalyst concentration, the FAME content peaked at catalyst concentration of 1.25 wt.% and at the concentration above that, the FAME content was lower than the maximum. In general, the conversion rate increases with catalyst concentrations; however, an excessive amount of catalyst can react with triglyceride, lowering conversion efficiency due to increased saponification [18, 37, 39].

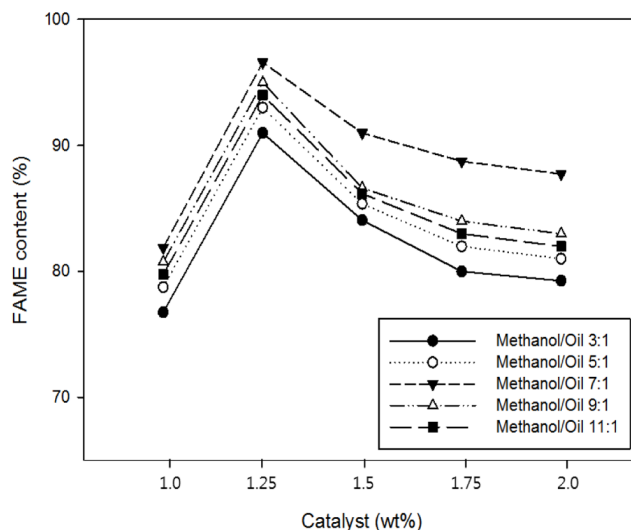


Fig. 3. Effect of molar ratio of methanol to oil and catalyst concentration on FAME contents at ultrasonic amplitude of 99.2 μm and ultrasound irradiation time (reaction time) of 10 min.

The FAME content in the BD produced using the optimal ultrasound-irradiation condition was 97.2% and the BD yield was 74%. In the meanwhile, the highest FAME content in the BD produced using mechanical string was 87.8% and the BD yield was 56.2%. Compared to mechanical transesterification, ultrasound-assisted transesterification improved the FAME content and BD yield by 9.4% and 17.8%, respectively. In addition, the reaction time was significantly shortened in ultrasound-assisted transesterification (10 vs. 60 min.) [14]. Together, the reduced amount of catalyst (by 37.5%) and reaction time (by 83.3%) could improve the economic efficiency of BD production from SCG. In addition, the BD production per hour was 106.8% higher when ultrasound irradiation was used instead of mechanical stirring. Regarding the FAME content, only BD produced using the ultrasound-assisted process satisfied the standard requirement. The effects of ultrasound irradiation on the FAME content in BDs have been reported in previous studies. Francisco et al. [40] reported that the ultrasound-assisted transesterification of fish waste oil increased the FAME content by 15% compared to the conventional mechanical stirring. According to Stavarache et al. [39], the ultrasound-assisted transesterification process in the presence of KOH as a catalyst resulted in a 33% reduction in reaction time and a 10% increase in the

FAME conversion compared to the mechanical stirring process. The ultrasonic cavitation generates energy to increase the interphase between oil and methanol, and consequently transesterification reactivity, allowing for a higher ester conversion in a reduced reaction time [14, 37].

3.4. Analysis of Energy Consumption

The energy consumptions as a function of the irradiated ultrasound amplitude are summarized in Table 2. The larger the amplitude, the higher the energy consumption. In the range of the amplitude between 24.8 and 117.8 μm , the ultrasound energy ranges from 5.28 to 362.9 kJ, and the power consumption from 17.6 to 100.6 W. The energy consumption in the best condition (amplitude 99.2 μm and reaction time 10 min) was 51.4 kJ. The amount of energy used in the transesterification reaction was 126 kJ. The amount of energy consumed by ultrasound-assisted transesterification was 59% lower than that by mechanical stirring. Compared to the ultrasound-assisted transesterification, the mechanical stirring process requires a longer reaction time and continuous heating to keep the reaction temperature constant, which could result in the overall higher energy consumption [41]. Yin *et al.* [41] reported that the energy consumed in the ultrasound-assisted transesterification of sunflower oil was approximately 42% lower than that of mechanical stirring. In their study, the reaction time was also reduced by 50% when ultrasound was used during

transesterification. Furthermore, ultrasound-assisted transesterification is reported to improve energy efficiency by approximately 45% compared to mechanical stirring [42]. As such, the energy requirement of the ultrasound-assisted process is lower than that of the mechanical stirring.

Table 3 shows the amount of energy and time required to produce 1 L of SCG-derived BD by ultrasound irradiation and mechanical stirring method. The ultrasound irradiation process was 12.5% lower in electricity consumption and 33.6% shorter in reaction time. The difference in reaction time can be explained with the ultrasound-induced reaction activation [43, 44]. Moreover, the mechanical stirring method requires hour-long heating, consuming a substantial amount of energy. According to Ji *et al.* [14], the ultrasound-assisted transesterification process of soybean oil achieved a higher FAME content within a shorter reaction time compared to the stirring method. In addition, the energy consumption for BD production was 250 Wh/kg for ultrasound and 500 Wh/Kg for mechanical stirring. The ultrasound-irradiation method consumed half of the energy required by the stirring method. Thanh *et al.* [45] reported that the FAME content increased from 81% to 99% with ultrasound irradiation. The additional benefit included less amounts of methanol and catalyst for transesterification, and reduced reaction time. These results show that the ultrasound-assisted conversion is an efficient method to produce BD in a short time with high energy efficiency [46].

Table 2. Amount of Irradiated Ultrasound Energy in Accordance with Ultrasound Amplitude and Irradiation Time (unit: kJ)

Irradiation time (min)	Ultrasound amplitude (μm)					
	24.8 μm	43.4 μm	62.0 μm	80.6 μm	99.2 μm	117.8 μm
5	5.3	9.7	15.1	19.4	25.8	30.1
10	10.5	19.5	30.2	38.8	51.4	60.4
20	21.1	38.9	60.2	77.7	102.8	120.7
30	31.8	58.3	90.4	116.4	154.3	181.0
40	42.7	77.8	120.8	155.3	205.7	240.9
50	52.5	97.2	150.6	193.8	257.0	301.8
60	63.4	116.6	181.0	232.9	308.5	362.9

Table 3. Energy and Time Consumption in Accordance with Transesterification Methods

Process	Ultrasound irradiation	Mechanical stirring
Transesterification	Power (W)	119
	Time (h)	0.16
	Energy consumption (Wh)	19
Washing	Power (W)	150
	Time (h)	0.5
	Energy consumption (Wh)	75
Drying	Power (W)	210
	Time (h)	1
	Energy consumption (Wh)	210
	Time total (h)	1.66
	Energy consumption total (Wh)	304
		347.5

3.5. Economic Feasibility

The energy and material costs per 1 L of BD from SCG are summarized in Table 4. The ultrasound-assisted process was 45% and 14% lower in energy and material cost, respectively, compared with the mechanical stirring method. The total cost was reduced by approximately 44%. The cost reduction could be attributed to the reduced amount of catalyst required for transesterification, a higher BD yield, and a shorter reaction time. Stavarache et al. [47] reported that, compared with mechanical stirring, ultrasound-assisted transesterification improved the FAME conversion rate by 18% and reduced the reaction time and catalyst requirement. According to Singh et al. [48], by replacing mechanical stirring with ultrasound irradiation, the reaction time of the transesterification of microalgae oil could be reduced from 5 h to 15 min and the FAME content could be increased from 91 to 98%. According to Sivaramakrishnan et al. [46], for BD production from microalgal oil, the reaction time and energy consumption required were 4 h and 38.09 kW, respectively, in the ultrasound-assisted process, and 36 h and 1,028.57 kW, respectively, in the mechanical stirring process. This suggests that the ultrasound-based BD production could be substantially more cost-effective. Overall, compared with the conventional mechanical stirring process, ultrasound-assisted transesterification is a more economically viable approach for BD production, featuring a shorter reaction time, lower energy consumption, and higher product purity.

In this study, BD was produced from untreated SCG using ultrasound-assisted transesterification reaction. In the mechanical stirring process, without pretreatment, the BD quality standard (FAME content ³ 96.5%) could not be met due to the high FFA content in SCG. For this reason, raw materials with high FFA contents

require a pretreatment process. To estimate the additional energy and material cost for SCG pretreatment, the SCG samples used for BD production were pretreated with sulfuric acid. The time and amount of energy required for the pretreatment process were 1.3 h and 0.14 kWh, respectively, which were translated into additional energy and material costs of US\$ 0.011/L and US\$ 0.029/L, respectively. According to Y. Zhang et al. [12], the cost for the pretreatment equipment accounts for 46% of the total equipment cost. When the energy and process time cost resulting from the pretreatment are added up to total process cost, the difference in economic efficiency between the mechanical stirring and the ultrasound-assisted process become more noticeable.

Overall, the ultrasound-assisted process could be an economically viable alternative to the conventional mechanical stirring method for a high-quality industrial BD production. The economic improvements are in part attributed to the direct use of SCG without any pretreatment.

3.6. Analysis of the Physical and Chemical Properties of SCG Biodiesel

The physicochemical properties of SCG BD are summarized in Table 5 in comparison with the BD and commercial diesel quality standards. The sulfur content, flash point, ash, water content, and density of SCG BD satisfied both quality standards. The oxidation stability of SCG BD was 6.6 h, higher than the BD standard of 6.0 h, and the cetane number was also higher than the BD standard. The cold filter plugging point (CFPP) of the SCG BD was -2°C, which was lower than the BD standard of 2°C, but higher than the commercial diesel standard of -18°C.

Table 4. Energy Cost and Used Material Cost in Accordance with BD Production Methods (unit: \$)

Process		Ultrasound irradiation	Mechanical stirring
Material cost	SCG	0.28	0.45
	Methanol	0.03	0.05
	Potassium hydroxide	0.07	0.19
Energy cost		0.024	0.028
Total		0.404	0.718

Table 5. Physicochemical Properties of SCG BD in Comparison with the Quality Standards of BD and Commercial Diesel

Specification	Unit	BD quality standard	Diesel quality standard	SCG BD
Sulfur content	mg/kg	< 10	< 10	8
Calorific value	MJ/kg	-	45	39.58
Flash point	°C	120 <	40 <	172
Ash content	(m/m) %	< 0.01	< 0.02	0.005
Water content	(m/m) %	< 0.05	< 0.02	0.015
Density at 15°C	kg/m ³	860 – 900	815 – 835	892.1
Oxidation stability	110°C, h	6 <	-	6.6
CFPP	°C	< 0	< -18	-2
Cetane number	-	47 <	52 <	48
Content of fatty acid methyl ester	Volume %	96.5 ≤	-	97.2

3.7. Analysis of Environmental Effect with SCG BD

BD is an environmentally friendly energy source (produced from biomass oils) that emits much less carbon monoxide (CO), hydrocarbon (HC), and particulate matter (PM) than conventional diesel (CD). According to a previous study, the PM, CO, and HC emission could be reduced by approximately 10, 21, and 11%, respectively, by adding BD to CD up to 20% [21]. Jalilantabar *et al.* [49] compared the soot, CO, and HC emission from engines running on CD and SCG BD20 (20% SCG BD blended with 80% CD by volume) at various engine loads. They found that soot, CO, and HC emission were down by 46%, 57%, and 77%, respectively, when the engine was fueled with SCG BD20. This could be associated with complete combustion facilitated by oxygenated components and lower sulfur and residual carbon contents in the BD [50-52]. Moreover, as a carbon-neutral fuel, the natural carbon sequestration reduces CO₂ emissions from BD by 50-80% compared to CD [53-55].

In Korea, the annual SCG production exceeds 120,000 tons (as of 2016). SCG is classified as industrial waste and is incinerated or used for landfill. The cost of SCG disposal is estimated to be approximately US\$ 13 million per year [56, 57]. According to previous report, the disposal cost of SCG can be as much as EUR€ 0.12/kg [58]. Landfilling and incineration of SCG have an adverse impact on the environment, including soil ecosystem destruction and greenhouse gas emissions [55]. The amount of GHG emissions from waste treatment in Korea (as of 2016) is 16.5 million tons of CO₂eq, which corresponds to 2.4% of the total greenhouse gas emissions [59]. SCG accounts for approximately 0.6% of all municipal waste, and the GHG emitted during the disposal is estimated at approximately 99,000 tons of CO₂eq. [57, 59, 60]. A kg SCG BD has a reduction effect of 2.1 kg CO₂eq, which means 21 tons of CO₂eq GHG can be saved by producing 10 tons of BD from SCG [31]. These analysis results show the environmental and economic benefits of using waste SCG [61].

4. Conclusions

In this study, we identified the optimal sonication parameters for the alkali-catalyzed transesterification of SCG oil. The highest FAME content (97.2%) was achieved at ultrasound amplitude of 99.2 μ m, reaction time of 10 min, methanol to oil molar ratio of 7:1, and KOH concentration of 1.25 wt.%. Compared with BD produced by the conventional mechanical stirring method, the FAME content of the SCG oil was increased, and the reaction time was reduced to one-sixth.

Due to the high FFA contents of the SCG oil, the BD produced through mechanical transesterification reaction did not meet the BD quality standard. In contrast, the ultrasound-assisted transesterification was efficient for producing the BD from untreated SCG oil that meets the BD standard. In addition, we found that energy and material cost can be reduced by replacing the mechanical stirring process with the ultrasound-assisted process. In conclusion, SCG-derived BD has substantial economic value with a small environmental footprint and could be a promising alternative to the current CD fuels.

Acknowledgments

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Author Contributions

Yeong-Su Kim (ph.D. student) conducted the experiments. Duk-Gam Woo (Post-doctoral researcher) as a corresponding author planned the experiments and wrote the manuscript. Tae-Han Kim (Professor) led the overall research as a corresponding author and helped revised the manuscript.

Nomenclature

AV	Acid Value
BD	Bio-Diesel
FAME	Fatty Acid Methyl Ester
FFA	Free Fatty Acid
LAME	Linolenic Acid Methyl Ester
SCG	Spent Coffee Grounds
KOH	Potassium hydroxide
NaOH	Sodium hydroxide
CFPP	Cold Filter Plugging Point
FID	Flame Ionization Detector
CD	Conventional Diesel

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