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Use of sewage sludge biochar as a catalyst in production of biodiesel through thermally induced transesterification

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Abstract

Sewage sludge (SS) is a residual/semi-solid material produced from industrial and municipal wastewater treatment processes. SS contains a high content of lipids and earth alkaline metals that can be used as catalysts for various chemical applications; however, its valorization has rarely been the focus of research. This study demonstrates that SS could be a promising raw material for biodiesel production and a biochar catalyst to promote the reaction kinetics of alkylation. Thermally induced transesterification of the SS extract (SSE) was performed in comparison with the conventional homogeneous reaction. SS biochar was fabricated via pyrolysis. The highest yield (33.5 wt.% per SSE) of biodiesel production was achieved in 1 min of reaction at 305 °C via thermally induced transesterification in the presence of SS biochar, while the yield of biodiesel from (trans)esterification with 5 wt.% H₂SO₄ was less than 1% even after 24 h. The reaction kinetics (< 1 min) of thermally induced transesterification was extraordinarily faster than that of conventional transesterification (3–24 h). The porous structure and high content of alkaline species in the SS biochar expedited the reaction kinetics. Consequently, the integrated/hybridized process for thermally induced transesterification and pyrolysis of the solid residue of SS was experimentally proved for the valorization of SS in this study. Considering that SS is being disposed of as a waste material and generates toxic chemicals in the environment, its valorization into value-added biodiesel and a catalyst could be an environmentally benign and sustainable technique.

Highlights

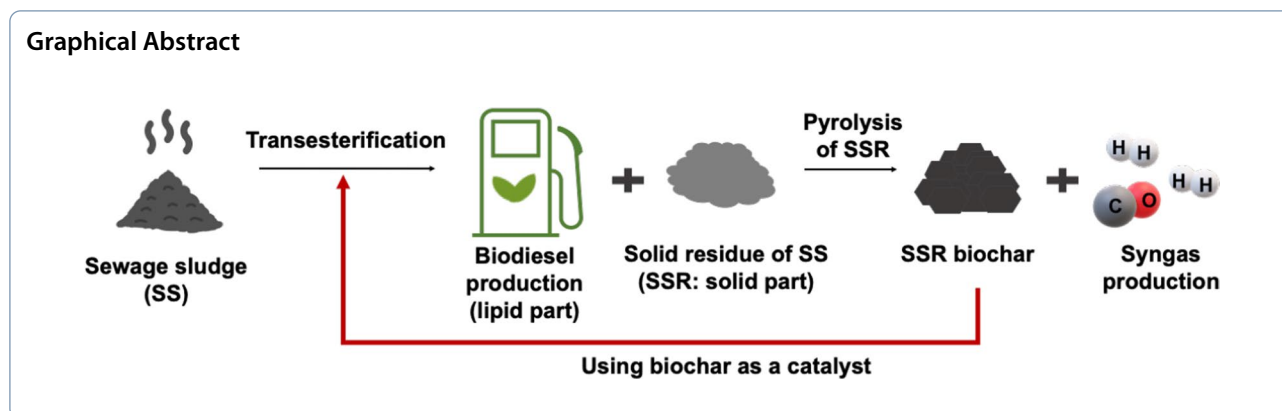
- Sewage sludge (SS) was converted into biodiesel and biochar catalyst.
- Thermally induced reaction using SS biochar had high biodiesel yield.
- SS biochar was more effective than porous silica in reaction kinetics.

Keywords: Circular economy, Waste-to-energy, Biochar catalyst, Biodiesel, Wastewater treatment

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1 Introduction

Biological wastewater treatment in wastewater treatment plants (WWTPs) harnesses the potential of bacteria and other microbes to consume organic nutrients via rapid metabolism (Ratola et al. 2012). As such, a large amount of sewage sludge (SS) is inevitably generated by WWTPs (Arhoun et al. 2019; Chang et al. 2020). This sludge is regarded as one of the main waste streams (Villalobos-Delgado et al. 2021), and it is estimated that more than 20 million tons of SS (dry/year) is generated worldwide (Melero et al. 2015). Composting, incineration, and landfilling are generally used for the disposal of SS (Mulchandani and Westerhoff 2016). Composting is a type of aerobic digestion that degrades biopolymers (fat, protein, hemicellulose and cellulose) in SS to be used as fertilizer (Anjum et al. 2016). Because composting is performed under mild conditions with natural airflow, the energy demand is low (Pergola et al. 2018); nonetheless, it is a lengthy process that lasts for a few months and requires a large area (Morsink-Georgali et al. 2022; Pergola et al. 2018). Unwanted control of nuisance (odor) and CO₂ emissions can also be problematic during the composting process (Chen et al. 2014). In addition, heavy metals and pathogens in SS composts have been regarded as problematic (Cai et al. 2007; Liu et al. 2017). SS incineration has been practiced for volume reduction (Fytilli and Zabaniotou 2008), but the desired air pollution control has not been readily achieved (Fytilli and Zabaniotou 2008). Specifically, the incomplete oxidation of SS arising from an uncontrollable ratio of SS to air could be a critical reason for the emission of hazardous pollutant mixtures such as polycyclic aromatic hydrocarbons (PAHs), dioxins, carbon monoxide, particulate matter (PM), and volatile organic compounds (VOCs) into the air (Moharir and Kumar 2019). Landfilling has been practiced as one of the main platforms for the

disposal of SS (Mulchandani and Westerhoff 2016), but it includes technical demerits such as large spaces, odor release, leachate generation, and long-term operation time (Ray et al. 2022; Syed-Hassan et al. 2017; Zhang et al. 2021).

In this context, it is of great importance to build a reliable and environmentally benign disposal platform for SS (Werle and Wilk 2010; Zabaniotou and Theofilou 2008). Adopting a pyrolysis platform as a practical disposal means could be a reasonable option to overcome these technical challenges. Pyrolysis is a thermochemical process that converts SS into the three-phased pyrolysates (char, biocrude, and syngas) through carbon rearrangement (reforming) under anoxic conditions (Bolan et al. 2022; Jung et al. 2019). As noted, SS can contain a large quantity of inorganics (such as metals) (Inguanzo et al. 2002; Jung et al. 2021b), and these materials can be immobilized by carbonization (biochar formation) (Lee et al. 2017b). Nevertheless, pyrolysis of the entire SS into three-phased products is not a viable option from an economic perspective because of a series of post-treatment process requirements of complex bio-oil compositions. It should be noted that SS contains up to 10–20 wt.% of lipid (dry basis) (Kargbo 2010; Manara and Zabaniotou 2012; Siddiquee and Rohani 2011a). Thus, the use of SS as a raw material in biodiesel production (fatty acid methyl esters (FAMES)) instead of lipid conversion into complex bio-oil could be a more cost-effective approach (Siddiquee and Rohani 2011b). Edible oils (soybean, rapeseed, sunflower, etc.) have been used as raw materials for biodiesel production (Hoekman et al. 2012; Jung et al. 2021d), and their cost accounts for 80% of the total production cost (Demirbas, 2007). The conversion of the oil in SS into biodiesel could be beneficial as SS lipids are inedible and waste-derived materials (Jung et al. 2020d; Olkiewicz et al. 2016). Furthermore, an effort to produce biodiesel from SS could offer an

opportunity to circumvent food security issues and ethical dilemmas. Biodiesel is used by blending with petrodiesel without any engine modifications so that its fuel properties are comparable to those of petrodiesel (Lim and Teong 2010). The use of biodiesel could offer an effective and strategic means to mitigate detrimental energy security and climate change effects (Demirbas 2009).

Biodiesel is produced from edible oils using an acid/base-catalyzed transesterification platform (Jung et al. 2020a; Mahlia et al. 2020). The conversion of SS oil into biodiesel via the catalytic transesterification process is not easily achieved because of the complex nature of SS oil (high content of di-/mono-glycerides, phospholipids, sterol, free fatty acids (FFAs), impurities, etc.) (Choi et al. 2019; Kwon et al. 2012a). For example, acid/base-catalyzed transesterification of SS oil leads to a saponification reaction and slow reaction kinetics owing to the high content of FFAs and impurities (Jung et al. 2020a). Also, acid/base catalyzed transesterification requires a washing process to remove homogeneous catalysts, generating 0.3 tons of wastewater per 1 ton of biodiesel production (Veljković et al. 2014). To realize the conversion of SS oil into biodiesel, catalytic transesterification in the presence of a solid catalyst has been proposed; however, its practical application has been hindered by the low yield of biodiesel (Kwon et al. 2012a; Lam et al. 2010). In addition, the production of biodiesel from SS oil under supercritical conditions (250–450 bar and 350–450 °C) was developed (Lee and Saka 2010; Liu et al. 2021), but its practical application has suffered from low economic viability due to harsh operational conditions (Kwon et al. 2013). Therefore, it is important to seek and develop a new biodiesel conversion platform. Indeed, non-catalytic (thermally induced) transesterification using porous materials has been reported in previous studies done by the authors (Jung et al. 2021c; Kwon et al. 2012a, b). It has also been reported that thermally induced transesterification has a high tolerance against impurities and FFAs (Jung et al. 2016, 2020c). This thermally induced production of biodiesel was realized at ambient pressure in a short reaction time (≤ 1 min) (Jung et al. 2018; Lee et al. 2017a). Despite these technical merits, the non-catalytic conversion of biodiesel requires a high thermal energy of ≥ 350 °C (Kim et al. 2022); thus, converting biodiesel from SS in a low-temperature regime to save energy is desirable.

Therefore, this study emphasizes the conversion of biodiesel from SS using a thermally induced platform. As a strategic means to lower the operational temperature and maximize waste valorization, biochar made from SS was used as a porous material and a catalyst in the thermally

induced transesterification of SS. Biochar (derived from biomass pyrolysis) has been highlighted in recent years owing to its broad applicability and availability (Lee et al. 2017b; Wang and Wang 2019) as well as its intrinsic physicochemical properties (large surface area, high porosity, long-term stability, and high content of earth alkaline metals), rendering it useful in agricultural, industrial, and energy sectors (Lee et al. 2017b; Oliveira et al. 2017; Tan et al. 2017). Recently, biochar has been evaluated as a green and cost-effective catalyst (Chen et al. 2020; Qian et al. 2015). For example, previous studies have reported that biochar can be used as a heterogeneous base catalyst in transesterification owing to its alkaline metal (oxide) content (K, Na, and Ca) (Balajii and Niju 2019; Daimary et al. 2022; Lee et al. 2017b; Orege et al. 2022). Lipids (free fatty acids and mono/di/triglycerides) were extracted from SS (SSE) using a solvent/solvent extraction method, which was then used as a raw material for thermally induced transesterification. Solid residue after lipid extraction of SS was pyrolyzed at 600 °C to produce biochar, which was then used as a catalyst for thermally induced transesterification. The effectiveness of biochar on biodiesel production was scrutinized as a function of the transesterification temperature (75–380 °C). The performance of SS biochar was compared with that of silica, a porous material previously used in thermally induced transesterification (Jung et al. 2021c; Kwon et al. 2012a, b).

2 Materials and methods

2.1 Sample collection and preparation

The SS was collected from the Jungnang water reclamation center (a sewage treatment plant in Seongdong-Gu, Seoul city, Korea). The wet SS collected from WWTP was dewatered in a centrifuge (Supra 22 k, Han-il), and the semi-dried SS was spread on a stainless-steel tray for drying at 105 °C for 2 d. The dried SS was pulverized using a ball-milling machine to obtain a powdered foam sample (< 1 mm).

2.2 Chemical reagents

Soybean oil was purchased from a local grocery market in Seoul, South Korea. Barium hydroxide (95.0%), silica (mean pore size: 6 nm, surface area: 175–225 m²g⁻¹), methanol (MeOH, $\geq 99.9\%$), phenolphthalein solution (0.5 wt.% in ethanol/water), and a standard solution of FAMES (37 components of C_{4–24} FAMES) were purchased from Sigma-Aldrich (USA). Potassium hydroxide (95.0%) was purchased from Samchun Chemicals (Korea). Chloroform ($\geq 99.5\%$), dichloromethane (DCM, $\geq 99.9\%$), isopropanol (99.8%), *n*-hexane ($\geq 99.0\%$), nitric acid (70.0%), and toluene (99.8%) were acquired from Daejung

Chemicals and Metals (Korea). N₂ and air gases (ultra-high purity) were purchased from Green Gas (Korea).

2.3 Solvent extraction

Lipids in the powdered SS were extracted for use as raw materials for biodiesel production using a solvent/solvent extraction method incorporating hexane or hexane mixtures: (1) hexane, (2) hexane-methanol mixture (1:1 by volume), (3) hexane-MeOH-chloroform (1:1:1), or (4) hexane-MeOH-DCM (1:1:1). Dried SS powder (10 g) and 200 mL of each solvent were loaded into a Soxhlet apparatus where the lipid extraction was performed at 80 °C for 24 h. Then, the SSE (a mixture of lipid and solvent soluble impurities) and solvent were moved into an evaporator (Büchi R-300, Switzerland). To separate the solvent, the rotary evaporator was operated at 80 °C for 3 h to evaporate volatile solvents from the solvent/extract mixture. The acid value (AV) of the SS was determined using a standard color-indicator titration method (ASTM D974-12). The SS residue (SSR) after SS extraction was collected from the Soxhlet apparatus and then dried at 105 °C to remove moisture and solvent. The SSR and SSE yields were measured gravimetrically.

2.4 Thermogravimetric analysis

Thermogravimetric analysis (TGA) was performed using a TGA instrument (STA 449-F5, Netzsch) to monitor the thermolytic patterns of SS, its extract (SSE), and solid residue (SSR). Ten milligrams of each sample were used for the TGA study, working from 35 to 900 °C at a constant heating rate (10 °C min⁻¹). For the TGA study, inert gas (N₂) or air was used as a carrier gas at a fixed flow rate (100 mL min⁻¹) automatically controlled using an embedded mass-flow controller. Prior to the TGA run for each sample, a reference test with a blank sample holder was performed to draw the background TGA curve. The background curve of the blank sample was used to compensate for any fluctuation in the TGA curve arising from buoyancy effects under dynamic temperature changes in the TGA study.

2.5 Biochar production from the SSR

SSR biochar (SSRB) was produced by pyrolysis under inert atmospheric (N₂) conditions. In detail, 20 g of SSR was placed at the center of the cylindrical quartz. The tube had an outer diameter of 60 mm, inner diameter of 56 mm, and length of 800 mm. The hollow furnace was covered with a cylindrical quartz tube to provide heat energy. The inner temperature of the cylindrical quartz was increased from 90 to 600 °C using an external furnace at a heating rate of 10 °C min⁻¹, and then pyrolysis was continuously performed at an isothermal

temperature of 600 °C for 4 h. Prior to pyrolysis, N₂ was flowed into the pyrolysis reactor for 30 min to remove air and then N₂ gas was continuously flowed until pyrolysis was completed.

2.6 Characterizations

Inductively coupled plasma-optical emission spectrometry (ICP-OES:720ES, Agilent) was used to determine the inorganic elements in the SSR and SSRB. The surface morphology of the SSRB was examined using field-emission scanning electron microscopy/energy dispersive X-ray spectroscopy (FE-SEM/EDX: Verios G4UC, Thermo Fisher Scientific). The crystal structures of the inorganic materials in the SSR and SSRB were examined using an X-ray diffractometer (XRD: D/Max-2500, Rigaku). Surface area, average pore volume, and pore size distribution of the SSRB were measured using a Brunauer–Emmett–Teller (BET) surface area instrument (BELSORP-mini II, Microtrac).

2.7 Biodiesel production

Biodiesel conversion from SSE was achieved using thermally induced transesterification. Conventional transesterification was performed as a reference using an acid catalyst. In the acid-catalyzed conventional transesterification, 10 g of SSE and 5 mL of MeOH were mixed in the presence of 5 wt.% H₂SO₄ in a 100 mL vial. The mixture was vigorously stirred with a magnetic stirrer (300 rpm) at 60 °C for 24 h. For thermally induced transesterification, closed batch-type reactors were used which were prepared using a stainless steel bulkhead union and two stoppers. A small-scale reactor was used to avoid heat transfer in the high-temperature range. One end of the bulkhead union was closed with a stopper and the reactants were added. The porous material (180 mg, silica or SSRB), 200 µL of MeOH, and 10 mg of SSE were added to the reactor in this order. Subsequently, 180 mg of porous material was added again before the other end of the reactor was closed with a stopper. The prepared batch samples were loaded into a muffle furnace to provide the thermal energy for the reaction. The inner temperature of the batch reactor was measured using a K-type thermocouple, and thermally induced transesterification was performed until the reactor reached the desired temperature (75–380 °C). The reactor was then removed from the muffle furnace and cooled to room temperature by circulating with cold water. The biodiesel produced from both conventional and thermally induced reactions was dissolved in DCM. The identification and quantification of the converted biodiesel were determined using multiple gas chromatography analyses. A schematic of the experimental setup is shown in Additional file 1: Fig. S1.

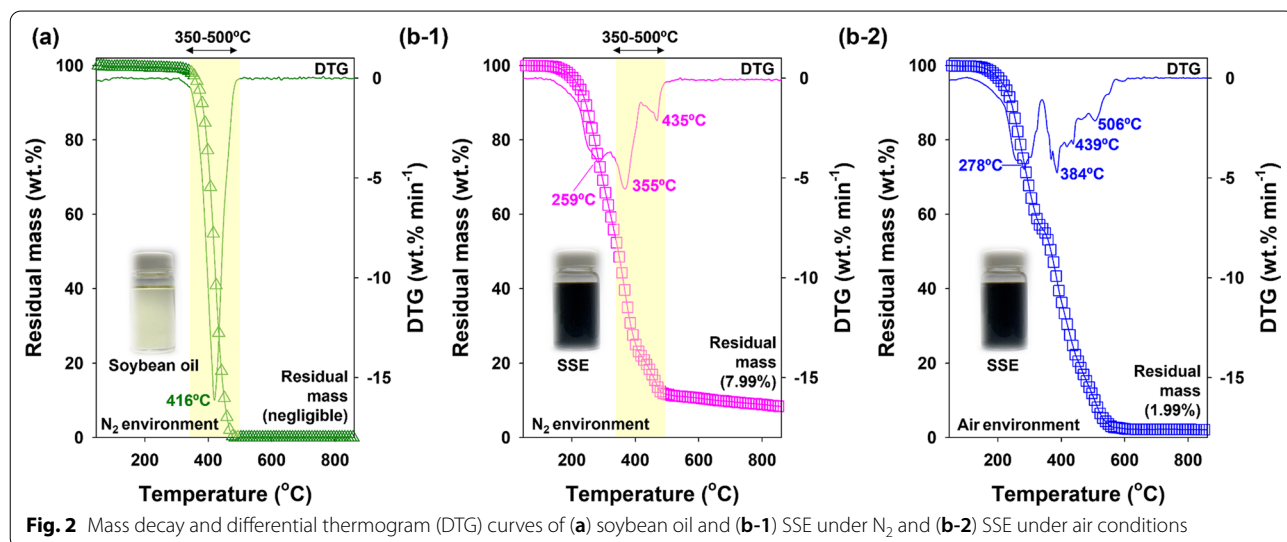
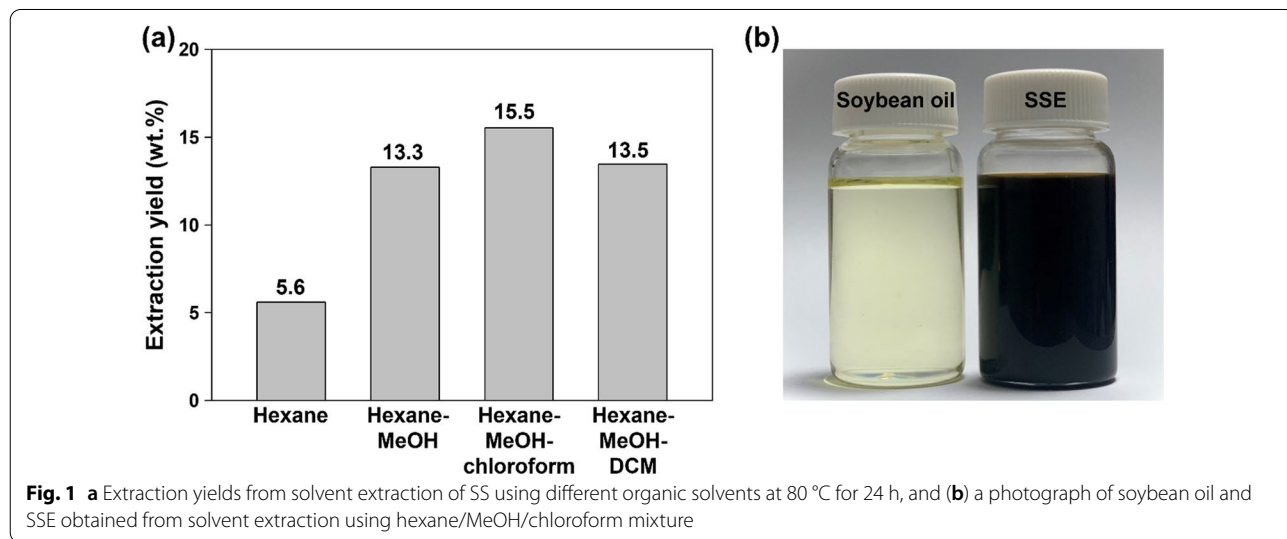
2.8 Product analysis

The major composition of biodiesel was determined using gas chromatography/time-of-flight mass spectrometry (GC/TOF-MS), and a gas chromatography/flame ionization detector (GC/FID) was used to quantify each biodiesel component. Both GC units were equipped with a DB-WAX column (0.25 mm (ID), 30 m (length), and 0.25 μm (film thickness)), and to calibrate the GC/FID system, a certified standard FAME mixture (37 FAME components) was used to construct the calibration curve. Detailed operating conditions of the GC/FID and all information on the calibration and quality assurance/control (QA/QC) of the FAME analysis are shown in Additional file 1: Tables S1 and S2–S5, respectively.

3 Results and discussion

3.1 Biodiesel production from SSE

Prior to upgrading the SS collected from the WWTP, valuable components were examined. The lipid content was experimentally determined using a solvent/solvent extraction method. Different solvent mixtures, such as hexane, hexane/MeOH (1/1 by volume), hexane/MeOH/chloroform (1/1/1 by volume), and hexane/MeOH/DCM (1/1/1 by volume) were utilized to maximize the extraction yield from the SS, as shown in Fig. 1a. The highest yield of the SSE (15.5 wt.%) was observed when the hexane/MeOH/chloroform mixture solvent was used. Unlike high-purity soybean oil, SSE is a dark brown solution containing undesired impurities (Fig. 1b). The AV of the SSE determined by a standard color-indicator titration



method was $63.7 \text{ mg KOH g}^{-1}$ SSE which indicates that the relative mass fraction of FFAs in SSE was 31.9 wt.% (Jung et al. 2020a). In contrast, the AV of high-purity soybean oil was lower than 1 because of the low fraction of FFA in it.

TGA tests of soybean oil and SSE were performed to quantify undesired impurities in the SSE. As shown in Fig. 2a, high-purity lipids had one sharp differential thermogram (DTG) peak at $417 \text{ }^\circ\text{C}$, and the thermal degradation of lipids was observed in the temperature range of $350\text{--}500 \text{ }^\circ\text{C}$. However, the SSE showed multiple DTG peaks and a wider thermal degradation window between 200 and $900 \text{ }^\circ\text{C}$ (Fig. 2b). The wide mass decay curve of SSE is ascribed to the thermal degradation of different types of organic compounds. For example, mass decay at temperatures $\leq 355 \text{ }^\circ\text{C}$ is responsible for the evaporation of volatile organic compounds and FFAs in SS (Cho et al. 2020), and the residual solid at $900 \text{ }^\circ\text{C}$ indicates the presence of soluble fixed carbon in SS. The ash component was rarely involved in the SS because the residual mass under air conditions was less than 2 wt.% (Fig. 2c).

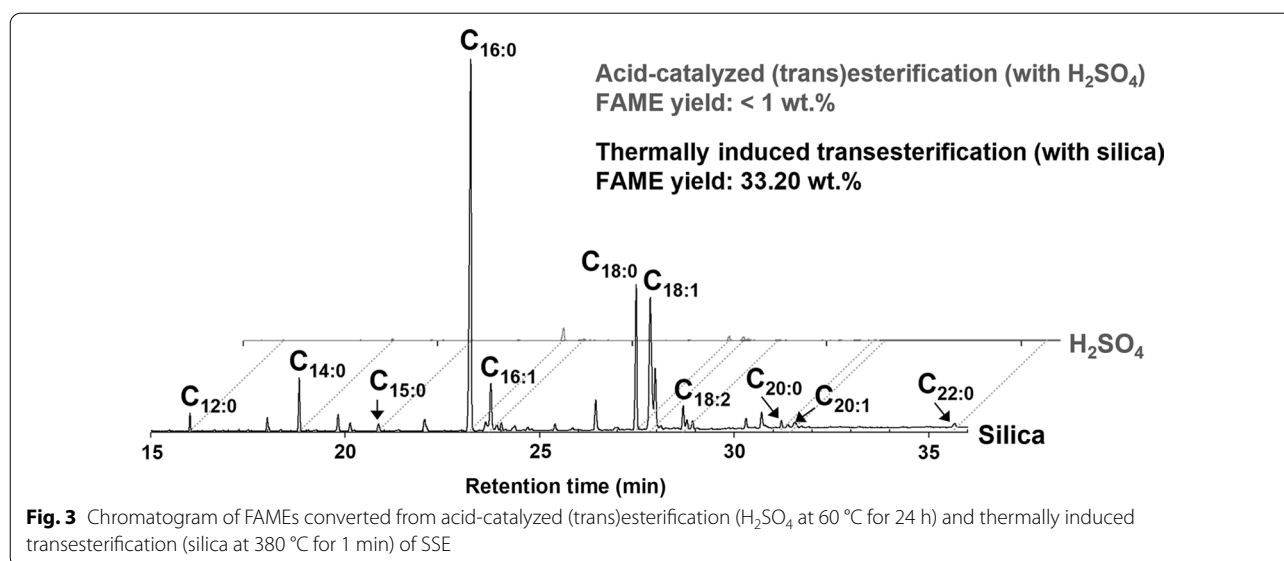
The SSE was converted into biodiesel using acid-catalyzed (trans)esterification to evaluate the effectiveness of conventional transesterification for this conversion. H_2SO_4 was chosen as the acid catalyst because the AV of SSE was much higher than 1 (Jung et al. 2020a). The biodiesel yield from acid-catalyzed transesterification (5 wt.% H_2SO_4) was less than 1 wt.% after 24 h of reaction at $60 \text{ }^\circ\text{C}$ (Fig. 3). This is plausible because the kinetics of (trans)esterification through conventional homogeneous reactions are highly sensitive to the presence of impurities (Choi et al. 2019; Kwon et al. 2012a).

In our previous studies, thermally induced transesterification was developed using porous materials (Jung et al. 2021c; Kwon et al. 2012a, b), which were invulnerable even in the presence of impurities such as water, organic compounds, FFAs, and inorganic species (Jung et al. 2016, 2020c). As such, thermally induced transesterification of SSE was performed using porous silica. The biodiesel yield from the thermally induced reaction was 33.2 wt.% after 1 min of reaction at $380 \text{ }^\circ\text{C}$, and the chromatogram profile of FAMES (biodiesel components) is displayed in Fig. 3. In our previous study, we also found that the biodiesel yield from low-quality lipids was higher than 95 wt.% (Jung et al. 2016, 2020c). However, the biodiesel yield from the SSE was 33.2 wt.%, meaning that SSE contains a variety of impurities as reported in the TGA result (Fig. 2).

3.2 Biochar catalyst production from pyrolysis of SSR

Thermally induced transesterification signified the conversion of SSE into biodiesel, whereas conventional acid-catalyzed (trans)esterification did not work for low-purity SSE. Nevertheless, the technical challenge in this process was the use of a higher reaction temperature ($380 \text{ }^\circ\text{C}$) to maximize the biodiesel yield when silica was used as a porous material.

Biochar derived from SS is a promising catalyst for thermally induced transesterification for several reasons. First, earth alkaline metals in organic waste can serve as catalytic materials to enhance the reaction kinetics of transesterification (Jung et al. 2020b); second, biochar has a porous structure (Gargiulo et al. 2018; Jung et al. 2019); and third, SS has a high potential to include silica and alumina from clay and sand (Díaz-Cruz et al.



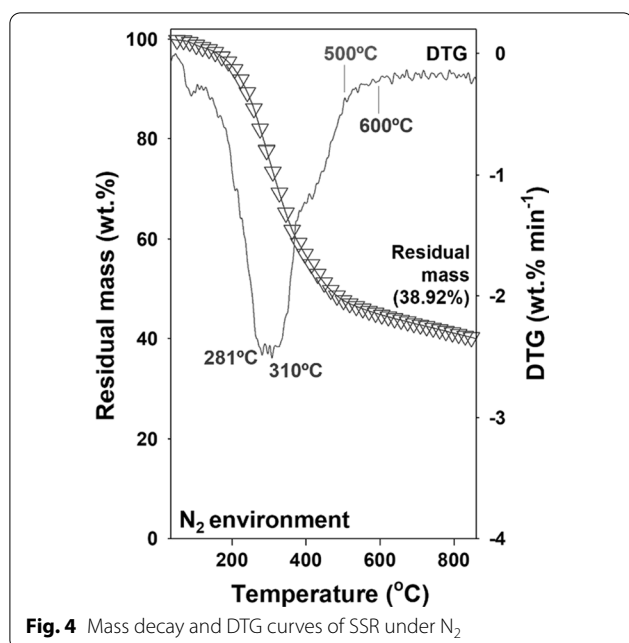


Fig. 4 Mass decay and DTG curves of SSR under N_2

2009) that could efficiently derive thermally induced transesterification.

To maximize the valorization of SS in the biodiesel production process, SS was converted into biochar and its characteristics were scrutinized. SSR (solid residue of SS after lipid extraction) was pyrolyzed to prepare the biochar (SSRB). To confirm the pyrolysis conditions for biochar formation, a TGA test of SSR was performed in the temperature range of 35–900 °C. As shown in Fig. 4, considerable mass decay of SSR was observed up to 500 °C. Rapid mass decay was observed due to the evaporation of volatile organic compounds in the SSR (Fig. 2). The mass decay was impeded at temperatures ≥ 500 °C and slowed at 600 °C. The slow reaction kinetics of the mass decay at 600 °C were attributed to the dehydrogenation and deoxygenation of the SS, which is called the carbonization process (biochar formation) (Liu et al. 2019). As the reaction temperature increased, more carbonization occurred, but the yield of biochar production decreased. To obtain a high biochar yield, the pyrolysis temperature for SSR was set to 600 °C, and the properties of SSRB produced at 600 °C were analyzed using multiple analytical tools.

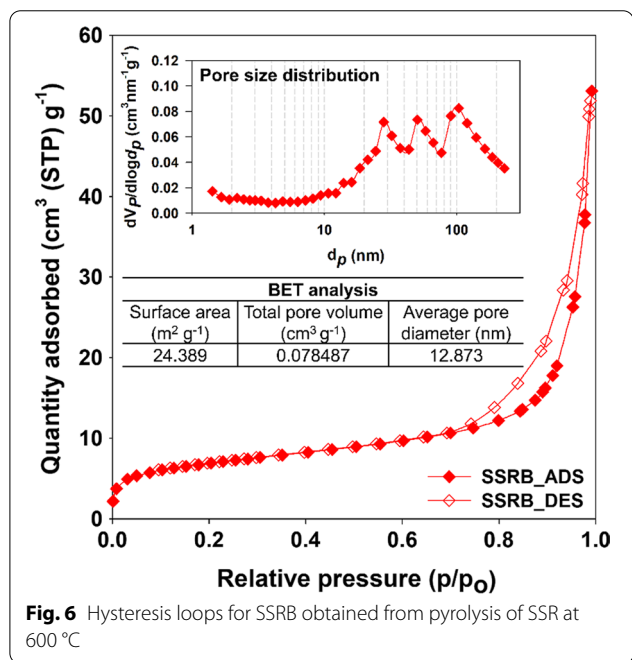
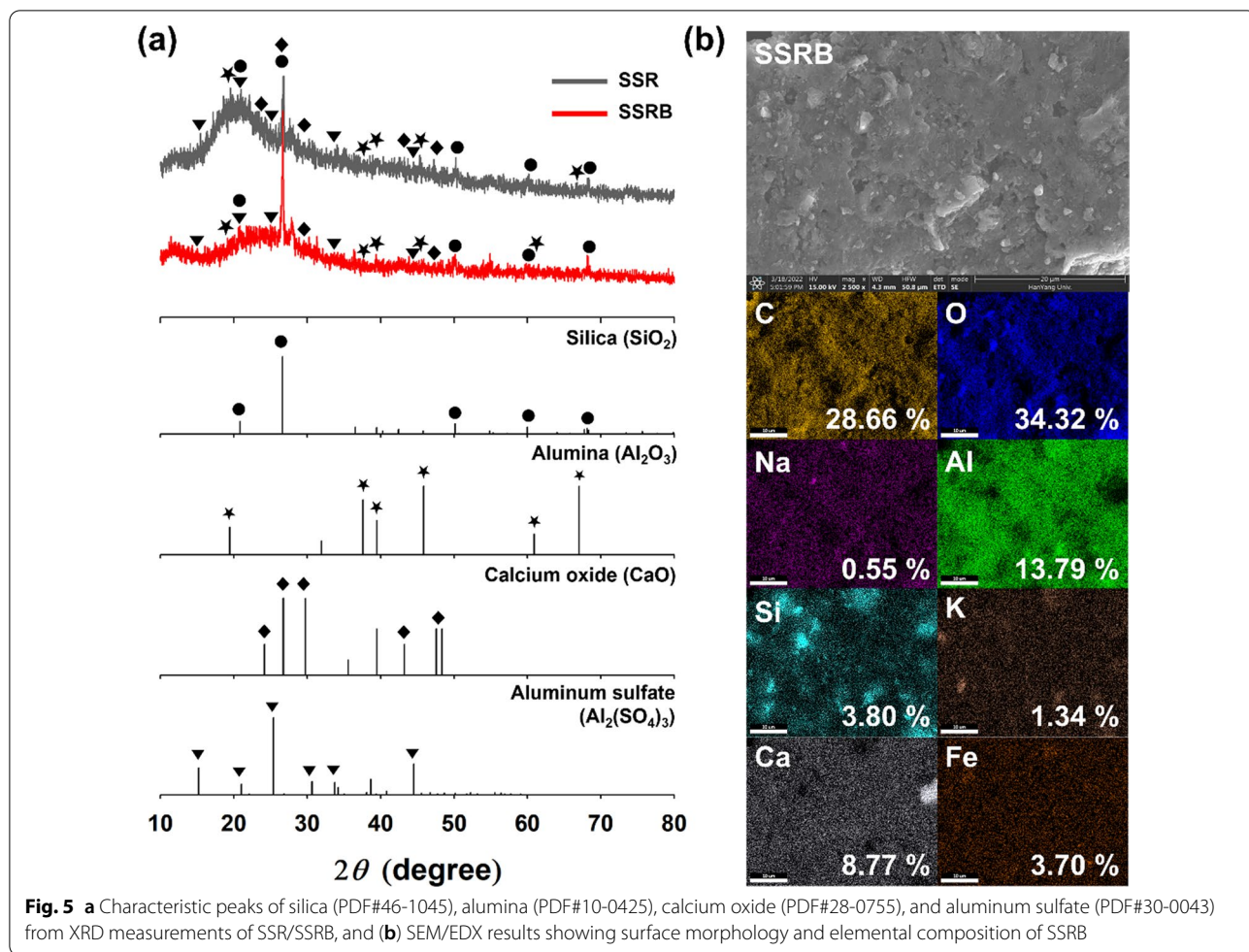
The inorganic contents in the SSRB were analyzed using ICP-OES. As expected, a variety of inorganic species were impregnated in the SSRB: Al (16.96 wt.%), Ca (4.06 wt.%), Fe (2.85 wt.%), K (0.75 wt.%), Mg (0.69 wt.%) and other trace materials (Table 1). When the SSR was converted into SSRB through pyrolysis, the inorganic content was enhanced by a factor of three. The results indicate that approximately two-thirds of volatile organic compounds in the SSR were converted into gaseous compounds through deoxygenation and dehydrogenation processes while fixed carbon was left in the form of biochar.

To further examine the detailed chemical structure of the inorganic species in the SSRB, XRD and SEM/EDX analyses were conducted. According to the XRD measurements, the major mineral component in the SSRB was silica (Fig. 5a) while alumina, calcium oxide, and aluminum sulfate were also observed. The presence of high levels of Al, Si, Ca, and oxygen was confirmed by the SEM/EDX analysis (Fig. 5b). In WWTPs, different types of coagulants are used to agglomerate solid waste materials, ultimately removing the settled agglomerates (Al-Mutairi et al. 2004). The most widely used coagulants are aluminum sulfate ($Al_2(SO_4)_3$), ferric sulfate ($Fe_2(SO_4)_3$), ferric chloride ($FeCl_3$), sodium aluminate ($NaAlO_2$), and calcium hydroxide ($Ca(OH)_2$) (Al-Mutairi et al. 2004; Song et al. 2004). Thus, the high content of inorganic species in WWTPs is mainly derived from coagulants. Because silica and alumina are the major components of sand and clay, it can be inferred that they are derived from sand and clay (Jung et al. 2021a). Calcium, potassium, and sodium are present in a variety of biomass waste materials.

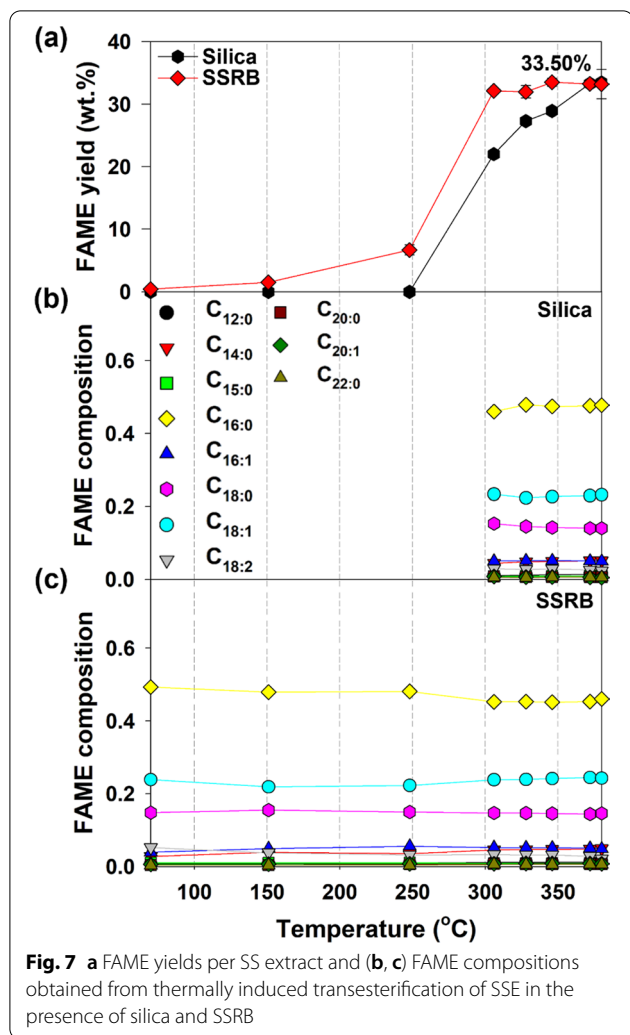
The porosity of SSRB was also examined using BET/BJH analyses, and the surface area ($24.389 \text{ m}^2 \text{ g}^{-1}$), total pore volume ($0.078487 \text{ cm}^3 \text{ g}^{-1}$), and average pore diameter (12.873 nm) were confirmed (Fig. 6). As shown in Fig. 3, silica was used as an effective porous material for thermally induced transesterification of SSE. The mean pore size and surface area of the silica were 6 nm and $175\text{--}225 \text{ m}^2 \text{ g}^{-1}$, respectively, while SSRB had a larger pore size and smaller surface area than silica. Although the porosity of SSRB was lower than that of silica, it was still acceptable for thermally induced transesterification as the average pore diameter of SSRB (12.873 nm), which

Table 1 Inorganic contents in the SSR and SSRB as measured by ICP-OES analysis

| Inorganic content (wt.%) | Al | Ca | Fe | K | Mg | Na | Zn | Cu | Ba | Ti | Total |
|--------------------------|-------|------|------|------|------|------|------|-------|-------|-------|-------|
| SSR | 5.12 | 1.68 | 1.25 | 0.32 | 0.32 | 0.15 | 0.09 | <0.01 | <0.01 | <0.01 | 8.93 |
| SSRB | 16.96 | 4.06 | 2.85 | 0.75 | 0.69 | 0.29 | 0.20 | 0.10 | 0.08 | 0.08 | 26.06 |



is larger than the molecular size of lipids (~2 nm), is indicative of a mesoporous structure. Note that thermally induced transesterification promotes chemical reactions between lipids and alcohols within the mesopores and macropores in porous materials according to our previous study (Kwon et al. 2012a). In the porous structure, the collision frequency between liquid- and gas-phase alcohols increases markedly, thereby leading to a rapid transesterification reaction. The required conditions for collision frequency enhancement are a mesoporous structure and porous material with a pore size larger than the diameters of lipids (monoglycerides, diglycerides, and triglycerides) and FFAs. As such, it was inferred that a larger SSRB diameter would enhance the collision frequency between lipids and MeOH. In addition to the porous structure, earth alkaline metals in SSRB are expected to enhance the reaction kinetics of thermally induced transesterification because they have been broadly used as solid catalysts for transesterification (Hoekman et al. 2012).



3.3 Biodiesel production from SSE using a SSRB catalyst

The effectiveness of earth alkaline metals and SSRB pores on thermally induced transesterification of SSE was examined by comparing the FAME yields when two different porous materials were used (silica vs. SSRB). Thermally induced transesterification was performed at a range of temperatures of 75–380 °C (Fig. 7).

The maximum FAME yields from both porous materials were higher than 33 wt.% at 380 °C (Fig. 6a), meaning that the porosity of SSRB was sufficient to drive thermally induced transesterification. An additional benefit of SSRB was the enhanced reaction kinetics of transesterification at lower temperature ranges. Specifically, the FAME yield from the thermally induced reaction of SSE with SSRB was greater than 32 wt.% at ≥ 305 °C, while it was less than 21 wt.% in the presence of silica at 305 °C (Fig. 7). The initiation temperature (150 °C) of the thermally induced transesterification with SSRB was also much lower than that (> 250 °C) with silica. The FAME

Table 2 FAME compositions obtained from thermally induced transesterification of SSE in the presence of silica and SSRB

| FAME component (carbon length: # of double bonds) | Silica | SSRB |
|---|--------------|--------------|
| Lauric acid (C12:0) | 1.45 ± 0.01 | 1.30 ± 0.01 |
| Myristic acid (C14:0) | 5.12 ± 0.05 | 4.94 ± 0.00 |
| Pentadecanoic acid (C15:0) | 0.97 ± 0.00 | 0.93 ± 0.00 |
| Palmitic acid (C16:0) | 47.75 ± 0.10 | 46.03 ± 0.01 |
| Palmitoleic acid (C16:1) | 4.97 ± 0.06 | 4.87 ± 0.02 |
| Stearic acid (C18:0) | 14.04 ± 0.02 | 14.64 ± 0.01 |
| Oleic acid (C18:1) | 23.18 ± 0.14 | 24.32 ± 0.02 |
| Linoleic acid (C18:2) | 2.34 ± 0.04 | 2.20 ± 0.00 |
| Arachidic acid (C20:0) | 0.61 ± 0.02 | 0.70 ± 0.01 |
| Eicosenoic acid (C20:1) | 0.47 ± 0.01 | 0.79 ± 0.00 |
| Behenic acid (C22:0) | 0.55 ± 0.01 | 0.59 ± 0.00 |

compositions were identical for both porous materials (Fig. 7; Table 2), indicating that SSRB significantly enhanced the reaction kinetics without altering FAME composition.

SSRB enhanced the reaction kinetics of thermally induced transesterification, although it was less porous than silica. The irregular porous structure of SSRB in reference to silica could be a disadvantage, but the high content of earth alkaline metals likely impart a catalytic capability, which could compensate for this. If the porosity is further controlled through additional activation processes, SSRB could exhibit better reaction kinetics in future studies.

In our previous studies, FAME yields from the thermally induced transesterification of high- and low-purity lipids were higher than 95 wt.%, regardless of the presence of impurities (Jung et al. 2016, 2020c). Considering that 95 wt.% of lipids in SSE were converted into biodiesel based on our previous results, it can be inferred that lipid content in SSE was 35.5 wt.%. As shown in Fig. 1, the SSE yield from solvent extraction was 15.5 wt.%, and the estimated lipid content in the dried SS was 5.47 wt.%.

The amount of lipids in SS can be highly susceptible to the types of waste materials in wastewater, as well as the weather, location, and other conditions (Bora et al. 2020; Edeh et al. 2019; Syed-Hassan et al. 2017). The SS used for thermally induced transesterification and biochar formation (Figs. 1, 2, 3, 4, 5, 6, 7) was collected during the winter season in Korea after heavy snowfall (December 2021). For comparison, thermally induced transesterification of SSE was also performed using samples collected during the dry summer season in Korea (August 2021).

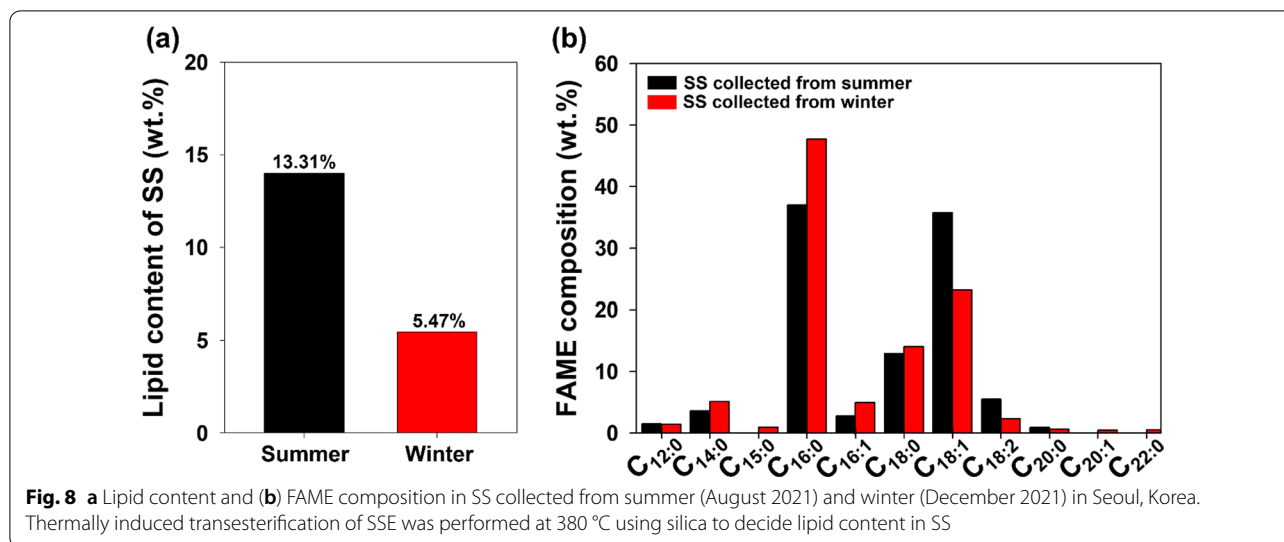


Figure 8 shows that SS collected in summer has a much higher lipid content (13.3 wt.%) than that from winter (4.9 wt.%).

The biodiesel yield and reaction kinetics investigated here were higher than those previously reported (Table 3). Multiple studies have reported that biodiesel yields from SS (trans)esterification were in the range of 0.4 to 13.3 wt.% (per dried SS). The considerable difference in biodiesel yield was ascribed to the different SS lipid fractions, conversion efficiency, and reaction kinetics. In the conventional (trans)esterification of SS and SSE using homogeneous catalysts (NaOH, KOH, and H₂SO₄), the conversion of lipids in SS to biodiesel was less than 40 wt.% after 5 to 24 h of reaction (Patiño et al. 2021; Patiño et al. 2018; Siddiquee and Rohani 2011a; Supaporn and Yeom 2016; Wu et al. 2017) (Table 3). The conversion efficiency and reaction kinetics were enhanced when solid catalysts were used. Higher than 42 wt.% of biodiesel yield was observed within 3 to 5 h of reaction time (Melero et al. 2015; Wu et al. 2017; Zhang et al. 2020).

From thermally induced transesterification using SSRB, a higher conversion efficiency (>95%) was observed after less than 1 min of transesterification reaction. Alkaline materials in the SSRB lowered the reaction temperature to reach the maximum yield of biodiesel from 380 °C (silica) to 305 °C (SSRB). However, the temperature required for thermally induced reactions is still higher than that required for transesterification reactions with homogeneous and solid catalysts. Thus, modification of SSRB to enable a lower reaction temperature could be a prospective study.

3.4 Estimation of biodiesel production from SS

To estimate the economic feasibility of biodiesel production from SS, the annual production of SS biodiesel was calculated based on the experimental results of this study (Fig. 8 and Table 3). According to the United States Northeast Biosolids Residuals Association (NEBRA) (North East Biosolid and Residuals Association 2007), the Ministry of Environment of Korea (South Korean Ministry of Environment 2019), and the Japan Statistics Bureau (Ministry of Land Infrastructure Transport and Tourism 2007), the annually generated amounts of SS in each of these countries were 7.18, 4.22, and 2.34 million ton, respectively (Table 4). Lipid content varies according to location, weather, and waste type, and as shown in Fig. 8 and Table 3, the available biodiesel yields from thermally induced transesterification were 5.47 wt.% and 13.31 wt.% during winter and summer, respectively. To calculate the estimated biodiesel yield from the SS, the average biodiesel yield (9.39 wt.%) was multiplied by the annual SS production in each country. From the calculation, it was expected that 0.65, 0.38, and 0.21 million ton of biodiesel could be produced from the U.S., Korea, and Japan, respectively, resulting in a financial benefit ranging from a few hundred million to one billion US dollars (Table 4). Considering that the global generation of SS is greater than 20 million tons per year, the financial benefit of biodiesel from SS could be higher than three billion US dollars annually.

This brief calculation does not reflect the capital and operating costs of the entire biodiesel production process, from SS collection to the biodiesel product delivery process. Therefore, a more detailed economic

Table 3 Comparison of conditions of biodiesel production from sewage sludge

| Sludge feedstock | Type of catalyst | MeOH to feedstock ratio ^a (by weight) | Catalyst loading per SS or lipid (wt.%) | Reaction condition | Biodiesel yield per lipid (wt.%) | Biodiesel yield per dried SS (wt.%) | Refs. |
|---|---|--|---|--------------------|----------------------------------|-------------------------------------|------------------------------|
| Homogeneous reaction | | | | | | | |
| Activated sludge | NaOH | 10 | 700 | 60 °C for 5 h | 14.3 | 2.92 | Patiño et al. (2021) |
| Activated sludge | H ₂ SO ₄ | 10 | 0.4 | 60 °C for 5 h | 22.2 | 4.53 | Patiño et al. (2021) |
| Sewage sludge | KOH | 10 | 30 | 60 °C for 5 h | – | 1.2 | Wu et al. (2017) |
| Floating sludge (extract) | H ₂ SO ₄ | 1.37 | 3.2 | 55 °C for 24 h | 26.8 | 0.40 | Patiño et al. (2018) |
| Floating sludge | H ₂ SO ₄ | 19 | 183 | 55 °C for 24 h | – | 2.10 | Patiño et al. (2018) |
| Primary sludge (extract) | H ₂ SO ₄ | 11.2 | 53 | 60 °C for 24 h | 38.9 | 5.63 | Siddiquee and Rohani (2011a) |
| Blended sludge (extract) | H ₂ SO ₄ | 2.3 | 2.9 | 70 °C for 8 h | 39.0 | 5.66 | Supaporn and Yeom (2016) |
| Heterogeneous reaction | | | | | | | |
| Sewage sludge | KOH/activated carbon | 10 | 3 | 60 °C for 5 h | - | 6.8 | Wu et al. (2017) |
| Primary sludge (extract) | Zr-SBA-15 | 0.69 | 1.7 | 209 °C for 3 h | 42.7 | 5.8 | Melero et al. (2015) |
| Sewage sludge (extract) | SO ₄ ²⁻ /Al ₂ O ₃ -SnO ₂ | 10.1 | 8 | 130 °C for 4 h | 73.3 | 11.80 | Zhang et al. (2020) |
| Thermally induced reaction (this study) | | | | | | | |
| Sewage sludge extract from winter | SSRB | 15.8 | 3600 | 305 °C for <0.01 h | > 95 | 5.47 | This study |
| Sewage sludge extract from winter | Silica | 15.8 | 3600 | 380 °C for <0.01 h | > 95 | 5.47 | This study |
| Sewage sludge extract from summer | Silica | 15.8 | 3600 | 380 °C for <0.01 h | > 95 | 13.31 | This study |

^aThe density of MeOH was assumed to be 0.792 g mL⁻¹

feasibility study as well as a life cycle assessment is required. The economic benefit of the SS upgrading process is highly contingent on the SS lipid content. Biodiesel production from SS lipids is an environmentally benign method with respect to the waste-to-energy concept, but the low lipid content in SS may not meet the economic feasibility of the process. Therefore, two strategic approaches are suggested to construct an efficient disposal and valorization platform for SS. When SS collected in winter has a low lipid content (e.g., 4.9 wt.%), direct pyrolysis of SS into value-added syngas, pyrolysis oil, and solid char could be more beneficial economically. When the SS

lipid content in summer is high (e.g., 13.3 wt.%), biodiesel production from SSE, following solid residue pyrolysis could be more beneficial. A schematic of the proposed SS valorization platform is shown in Fig. 9. SS is considered a waste material and is currently being discarded through incineration and reclamation, which is a costly but necessary process. Instead of disposal, this study suggests a new platform for the conversion of SS into biodiesel, biodiesel catalysts, and value-added syngas (industrial gas), which can simultaneously solve environmental issues and provide economic benefits.

Table 4 Economic assessment of biodiesel production from SS through thermally induced transesterification reactions

| | USA | KOREA | JAPAN |
|---|---|---|--|
| Annual generation of SS (dry basis) | 7.18 million ton (2004) (North East Biosolid and Residuals Association 2007) | 4.22 million ton (2019) (South Korean Ministry of Environment 2019) | 2.34 million ton (2019) (Ministry of land infrastructure transport and tourism 2007) |
| Biodiesel yield | 9.39 wt.% (dry basis) ^a | | |
| Estimated annual production of biodiesel (ton) | 674,202 | 396,258 | 219,726 |
| Current annual production of biodiesel in the U.S., Korea, and Japan ^b | 5.9 million ton (1.82 billion gallon) (U.S. Energy Information Administration 2020) | 650,000 ton (0.20 billion gallon) (Korean statistical information service 2019) | 20,000 ton (6.17 million gallon) (U.S. Department of Agriculture, 2021) |
| Portion of SS biodiesel to currently generated biodiesel | 11.43 wt.% | 60.96 wt.% | 1098.63 wt.% |
| Economic benefit from SS derived biodiesel (\$2.5 per gallon BD) | \$ 520 million | \$ 305 million | \$ 169 million |
| Economic benefit from SS derived biodiesel (\$5 per gallon BD) | \$ 1.04 billion | \$ 611 million | \$ 339 million |

^a Average biodiesel conversion yield from SS in summer and winter

^b 1 gallon of biodiesel converted to 3.244 kg

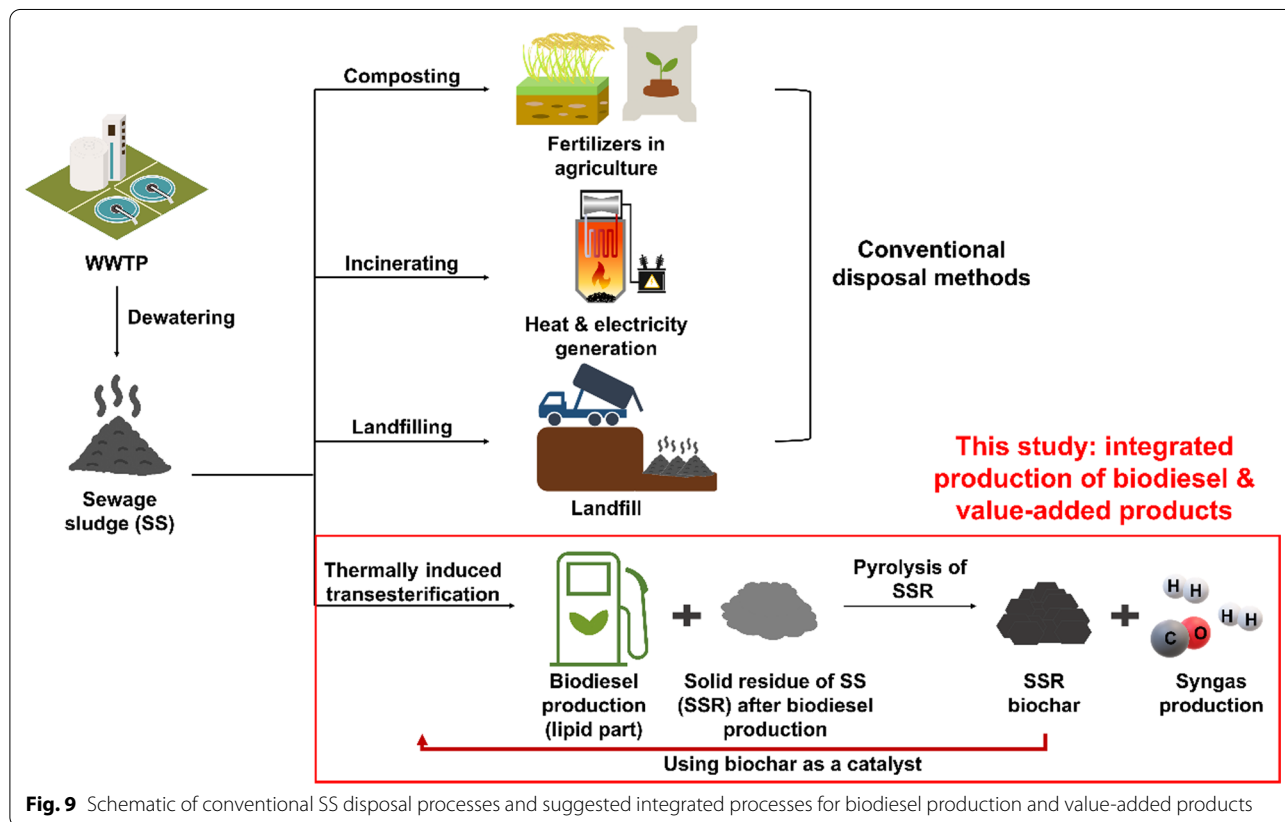


Fig. 9 Schematic of conventional SS disposal processes and suggested integrated processes for biodiesel production and value-added products

4 Conclusions

Here, SS was valorized into biodiesel and used as a catalyst for the biodiesel production process. The SSE, including lipids and organic impurities, was obtained using the solvent extraction method, and the SS solid residue was pyrolyzed to produce SSRB which has a

porous structure and high content of metallic compounds, such as Al, Ca, Fe, K, Mg, and Na. The synergistic effects of the porous structure and metallic compounds of SSRB promoted the reaction kinetics of thermally induced transesterification, resulting in a maximum yield of biodiesel (33.5 wt.% per SSE) at

305 °C within 1 min of the reaction. As a reference, thermally induced transesterification was performed using silica, a conventional porous material, where the required temperature for maximum biodiesel yield was higher (380 °C) because of the absence of catalytically active metallic species. The biodiesel yield and reaction kinetics of the thermally induced transesterification (33.5 wt.% at 305 °C for 1 min) were also much higher than those of acid-catalyzed transesterification with 5 wt.% H₂SO₄ (<1 wt.% at 60 °C for 24 h). This study proved that SS can be converted into value-added bio-fuel (biodiesel) and a catalyst to enhance the reaction kinetics of the biodiesel production process.

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1007/s42773-022-00194-7>.

Additional file 1: Fig. S1. Schematic of the experiment setups used in this study. **Table S1.** GC/FID operational conditions. **Table S2.** Concentration of FAME for 3-point RSE. **Table S3.** Results of 3-point RSE for QA/QC (Peak area). **Table S4.** Concentration of FAME for 7-point MDL. **Table S5.** Result of 3-point MDL for QA/QC.

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Author's contributions

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by SJ, MK, Y-HK, K-YAL, and EEK. Resources were provided from W-HC and YFT. All authors contributed to data curation. The first draft of the manuscript was written by SJ, MK, Y-HK and EEK, and all authors were involved in manuscript review and editing procedure. All authors read and approved the final manuscript.

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Availability of data and materials

Data available on request from the authors.

Declarations

Competing interests

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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