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1**Utilization of waste capiz shell – Based catalyst for the conversion of leather tanning waste into biodiesel**

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ARTICLE INFO ABSTRACT Editor: G.L. Dotto Rapid urbanization and technological advancement have led to worrisome challenges associated with increasing Keywords: waste production, and its management is overly burdensome. Indonesia annually produces 100,000 tons of Biodiesel leather tanning waste (LTW) and 2600 tons of waste capiz-shell (WCS). This study proposed a zero-waste ap- Catalyst proach by utilizing WCS as the catalyst for biodiesel production from LTW. Based on the characterization results, Waste capiz-shell the WCS-based catalyst is proven to possess high porosity and comparable catalytic activity to the other het- Leather tanning waste erogeneous catalysts. The maximum yield of FAEE was 93.4 wt%, obtained at 60 °C, 4 h reaction time, 3 wt% Zero-waste approach Viability study catalyst loading, and ethanol to LTW molar ratio of 6:1. High FAEE yield (> 90 wt%) can be obtained by reusing the WCS-based catalyst until the third reaction cycle. To examine the feasibility of this zero-waste act, a simple viability study was also performed by comparing this process to the conventional basic transesterification process. 1. Introduction Leather tanning and aquaculture industries are known to generate a higher amount of waste than products [1–3]. Approximately 80 wt% of the raw hide is generated as waste in leather processing [1,2], while more than 50 wt% of total production in the aquaculture industries is discharged as solid waste [3]. The financial and environmental costs for waste disposal from these industries can be burdensome and costly. Therefore, establishing a zero-waste act in these two industrial sectors to reduce the environmental impact is necessary through careful plan- ning, recycling, reusing, or composting as many as possible of the waste materials. Reusing and converting waste materials into highly valorized products is one way to conduct this zero-waste act. The valorization of biological sources to biodiesel has attracted extensive interests as it reduces the global petroleum demand and pollution [4,5]. Various types of feedstock, namely edible oils [6–8], non-edible oils [9,10], algae [11–13], industrial fats and greases [14–16],

3**have been** developed **to produce high-quality biodiesel using sundry of** transesterification **routes. The**

transesterification reaction is generally catalyzed by the presence of acids, bases, and enzymes, which can be added to the reaction system in the form of a homogenous or heterogeneous phase [17]. Currently, industrial-scale biodiesel pro- duction uses edible oil as raw material and NaOH or KOH solution in methanol as a homogenous alkali catalyst. However, high expenses of raw material and operation including the cost of homogenous catalyst and separation process become the major drawbacks in the use of biodiesel, particularly from the economic viewpoint [18]. Therefore, the use of leather tanning waste (LTW) as the raw material and waste capiz shell (WCS), one of the aquaculture wastes, as the catalyst source in biodiesel production may result in a declining operational ex- penditure. While the homogenous catalyst provides faster reactions and mild processing conditions [19], heterogeneous catalyst offers several ad- vantages over the homogenous ones, due to its reusability, easier Abbreviations: WCS, waste capiz-shell; LTW, leather tanning waste; RPO, refined palm oil;

1**FAEE, Fatty Acid Ethyl esters**; TG, triacylglyceride(s); **FFA, free fatty acid**

; CaO, calcium oxide ⁎ Corresponding authors. E-mail addresses: maria_yuliana_liauw@yahoo.com (M. Yuliana), sandy@ukwms.ac.id (S.B. Hartono).

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Received 22 March 2020; Received in revised form 29 April 2020; Accepted 1 May 2020 Available online 07 May 2020 2213-3437/ © 2020 Elsevier Ltd. All rights reserved. separation, and insensitivity to high contaminants, such as FFA, water and other minor components [20].

4**Furthermore, the regeneration of** heterogeneous catalyst **after the transesterification process is** easier **and produces** less **toxic wastewater**

, leading to a more environmentally friendly process [21]. Various heterogeneous catalysts for biodiesel production have been reported in literatures, including immobilized intracellular lipase [22], KOH/bentonite composite [23], sulphated zirconia [24,25], base/acid-supported resin [26,27], zeolites [25,28], metal-organic framework [29] and alkali/acid-impregnated meso- porous silica [30]. The

4**main problem for** the **utilization of these** het- erogeneous **catalysts is in** their environmental and economic values since

4most of these catalysts are synthetic, expensive, and difficult to prepare in large scale production [20]; which restrain their practical implementation in industrial scale

. Aside from the above mentioned heterogeneous catalysts, calcium oxide (CaO) has been widely investigated

5as a low-cost solid catalyst in biodiesel production

due to its abundant availability in nature and simple preparation [31–35]. CaO is also known to possess comparable catalytic activity to several homo- genous catalysts [36]. Limestone or seashells are common sources to produce CaO [31,33,34,37]. In Indonesia, the production of capiz-shell reached more than 4000 tons annually. The average weight of its shells is approximately 65 wt% and they are commonly discarded as waste [38]. Based on the data above, it is predicted that there will be a minimum of 2600 tons of WCS is annually produced. WCS contains high calcium content, reaching almost 20 wt% and can be used as an alternative source of CaO [39]. Currently, the utilization of WCS in the food sector has been so far reviewed by Agustini et al. (2011) to develop calcium-rich food based on WCS. Besides its wide studies in the food-related area, Suryaputra et al. (2013) studied the possibility of using WCS

4as raw material for new heterogeneous catalyst production in the biodiesel preparation

using refined palm oil (RPO). The maximum methyl ester yield (

493 ± 2.2 wt%) was obtained at 6 h reaction time and 3 wt% catalyst

loading, indicating that WCS

4has potential application as a renewable resource of CaO-based catalyst for biodiesel production

[20]. However, despite its high catalytic activity in converting refined oil to biodiesel, there has been no further study on the utilization of WCS as a heterogeneous catalyst source to convert a lipid-rich waste into biodiesel. In this study, LTW was co-used as the raw lipid material to complete the zero-waste act. According to the data provided by the Indonesian Ministry of Industry, around 100,000 tons of LTW is pro- duced annually in Indonesia, with a crude fat content of more than 60 wt% [1,40,41]. One of its valorization efforts has been converting the waste into biodiesel. Several studies have been performed to pro- duce LTW-based biodiesel using the following routes: base-catalyzed transesterification [14,42], solid-catalyzed transesterification using Cs2O loaded nano-magnetic particle [43], solid-state fermentation using micro bacterium species from soak liquor [44], and catalyst-free supercritical ethanol [41]. As the government plans to achieve sustainable development goals in 2030 which include affordable and clean energy,

maintaining cli- mate, and protect the ecosystem, the study on the zero-waste approach by exploiting these two waste materials (WCS and LTW) is an inter- esting

2**topic to be studied. The** focus **of this study is to observe the** potential use **of**

WCS as a catalyst for biodiesel production from LTW. The catalytic activity of WCS-based CaO was monitored at various operating parameters, namely reaction time, catalyst loading, and the molar ratio of LTW to ethanol. Ethanol was selected as the alcohol source instead of methanol, due to its sustainability since ethanol can be obtained from renewable sources [45,46]. The solubility of ethanol in oil will also increase the reaction rate and is favorable in a reaction system using a heterogeneous catalyst [46]. The recyclability of the WCS-based CaO was studied

3**at the operating condition giving the** maximum **yield**

. Moreover, a viability study has been also conducted to compare biodiesel preparation in this study with the conventional one.

2**2. Materials and methods 2.1. Materials** WCS **was collected from a local** fish market **in Gresik, Indonesia, and**

repeatedly rinsed to remove dirt and unwanted materials prior to pre- treatment,

2**following the procedure conducted by** Suryaputra **et al**

. (2013). The WCS-based catalyst powder obtained after the pre-treat- ment was then stored in a desiccator for further use. Meanwhile, LTW obtained from a leather tanning factory in Bogor, Indonesia was used as a lipid source for biodiesel preparation. Several pretreatment steps of LTW was performed before use according to the following procedures: LTW was washed three times with deionized water (1:1, w/w) to re- move gangue and other unwanted impurities. The water content in LTW was then removed by heating at a constant temperature of 120 °C and subsequently subjected to membrane filtration to obtain the treated LTW. Absolute ethanol and analytical grade n-hexane were purchased from Sigma-Aldrich and Merck (Germany), respectively. All chemicals used for the analysis were of high purity grade and require no further purification. The fatty acid ethyl esters (FAEEs) composition in the final biodiesel product were identified using an external standard pack (10008188) purchased

1**from Cayman Chemicals** (Ann-Arbor, **MI, USA), while methyl heptadecanoate** was used **as** an **internal standard** (IS) in **the** analysis **of FAEE**

purity. Nitrogen (99.99 %) and helium (99.9 %) gases for the

7**gas chromatography-flame ionization detector (GC-FID**) analysis **were**

provided

7**by Aneka Gas Industry Pty. Ltd., Surabaya. 2.2**

. Characterization of WCS based-CaO and LTW The characterization of WCS-based catalyst (WCS-based CaO)

3**was conducted using field emission scanning electron microscopy** (FESEM), **X-Ray**

powder diffraction (XRD) and thermogravimetric analysis (TGA). WCS-based CaO was analyzed for its morphologies using

2**FESEM JEOL JSM**-6500 F (**Jeol Ltd., Japan), with an accelerating voltage of** 15 **kV and**

12.4 mm working distance. The XRD pattern of the catalyst was acquired in the range of 15° to 90° (2θ angle) by using

2**an X'PERT Panalytical Pro X-Ray diffractometer (Philips-FEI, Netherlands) with monochromatic Cu Kα1 radiation at** a **wavelength (λ) =0.154 nm, 40 kV of voltage and 30 mA of tube current. The**

thermal stability of WCS-based CaO was studied

3**using TG/DTA Diamond instrument (Perkin Elmer, Japan**

). A 6-mg

6**sample was placed in a platinum pan and heated from**

30 °C

6**to 900 °C with a rate of 10 °C/min to monitor the** decomposition temperature **of the catalyst**

. To maintain the system O2- free, nitrogen with a velocity of 20 mL/min was purged into the system throughout the entire process. The determination of fat and FFA content, as well as the fatty acid composition in LTW as raw material for biodiesel preparation, were performed according to the standard methods of

1**AOAC 991.36, ASTM D5555-95 and**

ISO 12966, respectively.

6**GC-2014 (Shimadzu Ltd., Japan) equipped with Restek Rtx-65TG (30 m**

x0.25 mm ID x0.

1**10 µm film thickness, Restek, USA) as the** capillary **column** was used **to** measure **the** fatty acid composition **in LTW**

. Meanwhile, the molar

7**weight of** LTW **was** measured **using the equation** below: Molar **weight of**

$$LTW\left(MLTW , \right) = 56.1 \text{ x } 1000 \text{ x } g\ 3 \left(mol \right) (SV - AV)\ (1)$$

7**where SV is the saponification value of**

LTW (mmKOoiHl , mg/g)

7**and AV is the acid value of**

LTW (mmKoOiHl , mg/g) [47–49]. The characteristics of LTW are presented in Table 1. 2.3. The study of catalytic activity of WCS-based CaO in various transesterification conditions The catalytic activity of WCS-based CaO was determined by Table 1 The

3**characteristics of** LTW **as the raw material for biodiesel**

production. Parameters Result Water content, wt% FFA, wt%

1**Crude fat, wt% Molecular** weight (**g/mol**

) Fatty acid profile,

1**wt% C14:0 C16:0 C16:1 C17:0 C18:0 C18:1 C18:2 C18:3 C20:0**

13.48 14.20 65.82 878.5 2.08 30.71 4.03 0.25 16.42 44.59 5.95 1.65 0.27 conducting the catalytic transesterification of LTW to FAEE at various operating conditions. Three investigated parameters include the reac- tion time (h), catalyst loading (wt%),

1**and the molar ratio of ethanol to LTW. The** range **of** variation of the parameters was selected based on the study conducted by Suryaputra et al. (2013) and their relevance to the industrial feasibility, both in processing and economic viewpoint [20]. The procedure was carried out as follows: Ethanol and LTW at two molar ratios (6:1 and 12:1) were introduced to a three-neck round bottom flask equipped with a reflux condenser, mechanical stirrer, and heating mantle. A certain amount of WCS-based CaO (1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt% of LTW) was added to the system.

4**The mixture was** subsequently **heated at 60** °C **with continuous stirring**

1**at 700 rpm for** specified reaction time (**2, 3, 4 h). After the reaction completed, the** WCS-based CaO **was**

recovered by centrifugation and

4**re-calcined at 900** °C **for 2 h**, while the filtrate **was**

allowed to settle in the separatory funnel overnight to obtain two layers. The FAEE-rich phase (top layer) was then separated from the

4**bottom layer** consisted **of glycerol, excess methanol, and other**

by-products, before being subjected to vacuum evaporation for the excess methanol removal to obtain the final bio- diesel product. The analysis of FAEE purity and composition

2**was carried out using GC**-2014 (**Shimadzu, Japan), completely equipped with a split/splitless injector and a flame ionization detector (FID). The separation was performed using** the narrow bore **DB**

-WAX nonpolar capillary column (30 m x0.25 mm ID x0.25 μm film thickness,

2**Agilent Technology, CA). The column temperature was initially set at** 50 °**C**

and maintained iso- thermal for 15 min, before subsequently ramped to 220 °C at the rate of

7**4 °C/min. The** column **temperature was** then **held constant for another 15**

2**min. The temperature of the injector and detector were adjusted constant at** 250 °**C**

and 260 °C, respectively.

1**100 mg of** final **biodiesel product was dissolved in 2 mL of** internal standard **solution** (0.01 mg **ml**

−1) and

2**subjected to filtration using polyvinylidene** di- fluoride (**PVDF) filter prior analysis. The prepared sample (1 μl) was injected into the GC with a split ratio of 1:50. The velocity of nitrogen (N2, 99.9 %) as the carrier gas was fixed at 30 cm/s at 80 °C**

. The identification of FAEE composition in the final biodiesel product was conducted

3**by comparing the peaks in chromatogram with the external** FAEE **standard**

pack (10008188), while the purity of FAEE was calcu- lated using the following equation: FAEE Purity (Fp, wt%) = $\left(\left(\frac{\sum A_{FAEE} - A_{IS}}{A_{IS}} \right) \times \frac{V_{IS} C_{IS}}{m} \right) \times 100\%$ (2) Where ∑ A FAEE is the sum of the area of FAEE peaks, AIS is the cor- responding peak area of IS, VIS

3**is the volume of** IS **solution (ml**), CIS **is the concentration of** IS **solution (g/mL), m is the actual** sample weight **of the final** biodiesel **product (g**

). According to the Fp measured from Eq. (2), the determination of FAEE yield can be performed using Eq. (3): FAEE Yield (wt%) = $\left(\frac{m_{BD} \times F_p}{m_{LTW}}\right) \times 100\%$ (3) Where mBD

6**is the weight of final biodiesel** product (**g**

), mLTW

1**is the initial** weight **of LTW (g) and** Fp **is the FAEE purity** (wt%) **obtained from** Eq. (2). **2**.4. Recyclability study **of**

WCS-based CaO To measure the catalyst resistance to deactivation, the recyclability of WCS-based CaO was determined by reusing the catalyst

3**for the transesterification process at the condition** giving **the maximum yield of** FAEE. **The**

transesterification product was then analyzed for its purity and yield using GC-FID according to the procedure mentioned in Section 2.4. The repetitive transesterification process was conducted

1**using the same catalyst until the** FAEE **yield** obtained was **below 90 wt** % with **the** number **of**

repetitions regarded as the recyclability number of WCS-

1**based CaO. All** experiments **were** carried out **in triplicates**

.

4**3. Results and** discussions **3.1. Characterization of** WCS-based **CaO The** surface morphologies **of** natural **shell**, WCS, **and**

the treated WCS-

1**based CaO were captured by** using **FESEM**

, as shown in Fig. 1(a) and (b), respectively. It can be seen from the corresponding figure that the structure of natural WCS is changed significantly from smooth surface and lumpy architecture to a more powdery structure. Fig. 1(b) showed that the CaO catalyst obtained from the calcination treatment of WCS at 900 °C has a honeycomb-like porous and rough surface. The porous structure

5**is probably due to the fact that a large number of** bound **water** in the **molecular** state is released **from the**

WCS-based CaO during the calcination, creates high porosity in the catalyst [50]. This result is in agreement with the studies conducted by Hu et al. (2011) and Niju et al. (2014) for the treated egg-shells and mussel shells, re- spectively [50,51]. Some particles seem to be entangled to each other, causing aggregation on the particle and non-uniformity particle size. This phenomenon is likely due to the presence of pure carbon in the WCS, as shown in the EDX pattern (Fig. 1(c)), which was oxidized to carbon dioxide during the calcination and reacted with CaO to form $CaCO_3$ which is the main trigger of the agglomeration [52]. As seen from the BET data which was summarized in Table 2, the specific surface area and pore volume between WCS and WCS-based CaO catalyst also obviously differ each other, which was attributed to the calcination process of the catalyst. WCS-based CaO catalyst pos- sesses 10-folds larger surface area and 5-times higher porosity as compared to the untreated WCS, indicating the occurrence of impurities and bound water removal during the heat-activation treatment which play a vital role in improving the two important parameters related to the porosity of a catalyst. High

1**surface area and pore volume of** a solid **catalyst have a** direct and **proportional** impact **on its catalytic activity**

[35]. The experimental results showed that the surface area of WCS- based CaO obtained in this study is in the range of the two studies performed by Hu et al. (2011) and Niju et al. (2014), representing that WCS-

based CaO has the comparable ability as a catalyst for biodiesel production [50,51]. The WCS-based CaO was further analyzed by XRD and its diffraction pattern is shown in

1**Fig. 1(d). The diffraction pattern of** the WCS-**based**

catalyst is in accordance with the characteristics of CaO (JCPDS card NO. 82-1691) and calcite (JCPDS card NO. 29-0306), with CaO as the major component and the remaining calcite after decomposition as the minor phase. The thermal stability of the catalyst was observed from the TGA curve, shown in Fig. 1(e). Only one major weight-loss peak was identified, with a weight loss of around 5% at 571–736 °C, which corresponds to the removal of chemisorbed water [53], decomposition of CaCO3 and its phase transition to CaO [50]. This result is consistent Fig. 1. The characterization results of WCS and WCS-based CaO after calcination at 900 °C for 2 h: (a) SEM image of WCS, (b) SEM image of WCS-based CaO, (c) energy dispersive X-Ray (EDX) spectra of WCS, (d) diffraction pattern of WCS-based CaO, and (e) thermogravimetric analysis (TGA) curve of WCS-based CaO. with that previously reported by Tang

2**et al**. (2013) **and Zhu et al**. (2011), which reported **that the**

weight remains almost constant after the decomposition of calcite completed in the temperature of around 700 °C [53,54]. Based on the TGA results, it can be concluded that a temperature of 900 °C is suitable for the calcination since it decomposes almost all the calcite and its base derivatives (calcium hydroxide) and forms the porous structure which is desirable for a catalyst. Hu et al. (2011) mentioned that

5**higher activation temperature is** also **required to** escalate **the activity of** a solid **catalyst**

[50]. Table 2 BET surface area and porosity of WCS and WCS-based CaO. Materials SBET (m2 g−1) Vm (cm3 g−1) WCS 0.82 0.011 WCS-based CaO 8.49 0.049 3.2. Transesterification of

1**LTW to biodiesel using** WCS-**based CaO**

as a catalyst Table 1 presented the

3**characteristics of** LTW **as the raw material for biodiesel preparation**

, and a quite substantial amount of lipid was monitored in LTW, with a value of 80.02 wt%. Among those percen- tages, the FFA value covers around 14.20 wt% of the total mass of LTW, leaving around 65.82 wt% of crude fat including TG, other acyl gly- cerides, and minor lipid compounds. The GC chromatogram verified that the major fatty acids constituting LTW are

1**palmitic acid (C16:0), stearic acid (C18:0), and oleic acid (C18:1**). Looking at **the**

FFA value, the traditional conversion of LTW to FAEE using homogenous catalyst would require at least two processing steps: (1)

3**esterification to lower the FFA** value **by converting them into**

FAEE in the presence of acid catalyst and (2) transesterification to convert the other lipid components into biodiesel using a basic type of catalyst, due to the sensitivity of homogenous catalyst to the presence of impurities, e.g. water and FFA. On the other hand, heterogeneous catalyst shows the insensitive characteristics to the high content of water and FFA in the system [55], making it possible to reach a high yield of conversion using only one-step process. Fig. 2(a)–(b) summarized the FAEE yield obtained at various reac- tion time, catalyst loading, and the molar ratio of LTW to ethanol. Based on the experimental results, the maximum FAEE yield (93.4

1**wt%) with the purity of** 97.8 **wt**

% was obtained at the following conditions: 60 °C, 4 h, 3 wt% catalyst loading,

1**and a molar ratio of ethanol to LTW**

6:1. It can be seen from the figure that the catalyst loading has an overall

6**positive effect on the** FAEE **yield. The**

experimental results demon- strated that the increase of catalyst

6**loading from 1 wt% to** 3 **wt% greatly** escalates **the** FAEE **yield by**

1.5 folds in all conditions. WCS- based CaO plays its catalytic function as the alkaline active sites gen- erated from the surface excited the proton from ethanol to form calcium ethylate and hydroxyl ion. The calcium ethylate formed then acts as a nucleophile attacking the carbonyl carbon in the FFA and acyl glycer- ides chain, leading to the initiation of base-catalyzed reaction to form FAEE and glycerol as a by-product. Once a substantial amount of gly- cerol was produced, the existing WCS-based CaO in the system reacted with glycerol to produce calcium glyceroxide [56,57] which further reacted with the excess ethanol to form C2H5O-Ca-O(OH)2C3H5. This component deactivates the catalytic activity of CaO since it possesses lower basic strength. Therefore, further addition of catalyst loading from 3 wt% to 5 wt% gave an unfavorable influence to the yield of FAEE as it declines from ~90 wt% to around 70 wt% [20,57].

3**Wei et al. (2009) also mentioned that adsorption and desorption of reactants from the catalyst is the** actual **rate-determining step in the overall reaction** [58]. **Therefore**

,

5**adding more catalyst is not favorable** to **increase** the yield **of**

FAEE. The effect of reaction duration was investigated at three different levels from 2 h to 4 h. Fig. 2 presented that longer duration of reaction promoted a moderate enhancement of the FAEE yield in the constant value of ethanol to LTW molar ratio and catalyst loading. Longer con- tact between the reactants (alcohol and lipids) and catalyst

7**ensures the conversion of triglycerides and FFA into**

FAEE. However, its sig- nificance is not comparable to the effect of catalyst loading. Another key parameter in the process is the molar ratio of ethanol to LTW. Theoretically, the

5**stoichiometric molar ratio of** ethanol **to** LTW **for** biodiesel production **is 3:1**. However, **in practice, the ratio should be higher in order to** purposely **drive the reaction**

toward the product side Fig. 2. The FAEE yield (

6**wt%) based on the experimental results**

with the in- teraction between

6**catalyst loading (wt%) and time (h**

) at ethanol to LTW molar ratio of (a) 6:1 and (b) 12:1. and gain high product yield, since transesterification itself is a re- versible reaction [18,59]. As described in Fig. 2(a)–(b), the influence of ethanol to LTW molar ratio gave beneficial results on the FAEE yield at the lower half of catalyst loading (1–2 wt%). Excessive alcohol is de- sirable to ensure full contact between reactants and catalyst to accel- erate the rate of reaction. However, a lower

1**molar ratio of ethanol to LTW**

(6:1) gave a more

1**favorable effect on the yield of** FAEE when **the**

catalyst loading used is 3–5 wt%. It is likely due to the excess of me- thanol caused the rapid formation of glycerol which will drive the re- action back towards the reactant side, causing lower FAEE yield. Hu et al. (2011) reported similar phenomena in their study that excess alcohol to lipid

7**ratio seems to be favorable** to **the** FAEE **yield** only **to a certain extent**

and reaches a stagnant line thereafter [50]. As a matter of fact, the higher molar ratio of ethanol to LTW will only escalate the raw material cost and consume larger amounts of energy for the purification and rectification processes [18]. 3.3. Recyclability of WCS-based CaO One of the most important purposes of

4**using heterogeneous catalyst for biodiesel** conversion **is** the recyclability **of** a **catalyst**

. For the pur- pose of determining the recyclability of WCS-based CaO, subsequent reaction cycles were performed, with the following operating condi- tions: reaction temperature of 60 °C, 4 h reaction time, 3 wt% catalyst loading, and the molar ratio of ethanol to LTW 6:1. After each cycle, the solid catalyst

3**was recovered following the method** stated **in Section 2.4, while fresh** reactants **were used in every cycle. The**

experimental results are depicted in Fig. 3. The results indicated that high FAEE yield of FAEE yield, (wt%) 100 95 90 85 80 75 70 65 60 0 1 2 3 4 5 Transesterification cycle Fig. 3. The recyclability of WCS-based CaO as the catalyst for biodiesel pre- paration from LTW at the transesterification condition of 60 °C, 4 h, catalyst loading of 3 wt% and ethanol to LTW molar ratio 6:1. above 90 wt% was achieved until the third run with the purity of FAEE ranges from 97.1 to 97.8 wt%. However, the conversion of FAEE de- clines rapidly in the fourth run, reaching only 64.4 wt% of FAEE yield. The catalytic deactivation of CaO is generally

3**caused by the** direct **contact between** basic **sites on the catalyst surface and the deactivation- induced components**

[36]. Gaseous CO2 and water vapor in the ambient air form CaCO3 and Ca(OH)2 when in contact with the oxide catalyst, leading to a decrease in the surface area of the catalyst and subse- quently reduce the catalytic activity of CaO [56]. Moreover, as pre- viously mentioned above, the presence of glycerol in the reaction mixture drives the formation of a less active catalyst, C2H5O-Ca-O (OH)2C3H5. High FFA content in LTW also plays a major role in the deactivation of CaO catalyst because the acidic

3**FFA tends to neutralize basic sites in the**

surface, resulting in the generation of calcium-car- boxylate, a component that is miscible in the lipid phase and induces the formation of ethanol-oil emulsion [36]. The fuel characteristics

1**of LTW-based biodiesel** produced **using** WCS-**based CaO as** the **catalyst**

and its comparison to the ASTM Standards D6751−19 are reported

3**in Table** 3. **The measurement results** indicated **that**

LTW-based biodiesel possesses a

3**comparable combustion and flow properties** to **the** standard required. High flash point also showed that LTW-based biodiesel can be safely handled, stored, and transported. The cloud point of LTW-based biodiesel was found to be 10.1 °C, which is lower than that required by Indonesian National Standard (SNI 7182:2015, < 18 °C), but still considerably high in countries with severe winter. Several techniques can be industrially applied to improve the cloud point of biodiesel, including cold filtration [60], adsorption using various natural and synthetic sorbent materials [61,62] as well as the addition of cold flow improvers, namely olefin- ester copolymer, poly-methyl acrylate and ethylene-vinyl acetate co- polymer [63]. 3.4. Simple viability study of the utilization of WCS-based CaO as a catalyst

5**for the biodiesel production** from LTW **To** highlight **the** possible **utilization of**

WCS as the catalyst to pro- duce biodiesel from LTW, particularly in Indonesia, a simple viability study was performed. Taking into account the rapid growth of renew- able energy consumption and its competitive situation, the non-edible oil including LTW was preferred over the edible ones. Moreover, it is a good addition to use the widely developed heterogeneous catalysts generated from waste-origin. As mentioned, approximately 2600 tons of WCS [38] and 100,000 tons of LTW [40,41] were annually produced. The market price of WCS in Indonesia costs around 900 US$ per tons, while in the case of LTW, this type of waste is generally discarded to the environ- ment. Looking at the low market price, the two components possess a high possibility to be used as a replacement for the current edible oils and homogenous catalysts to produce biodiesel. Based on the experi- mental results, every 100 g of LTW contains lipid fraction of 80.02 g that can be converted into FAEE with the maximum yield of 93.4 wt% (74.74 g)

3**at the following conditions**: temperature of **60 °C**, reaction time of **4 h**

, catalyst loading of 3

1**wt% and molar ratio of ethanol to LTW** of 6:**1**. Therefore, **the**

annual production of biodiesel from LTW is able to reach 74,740 tons and fulfill approximately 1.0 wt% of the total biodiesel demand in Indonesia [64], with the total WCS-based catalyst and ethanol amount of 2,242.2 tons (3 wt% of LTW) and 25,194.7 tons (6:1 ethanol to LTW molar ratio), respectively. Our viability study compared the proposed technique of using WCS- based CaO as the catalyst to convert LTW into biodiesel (process A) to the conventional base transesterification using a homogenous catalyst to convert RPO into biodiesel (process B) and considered that the processing steps between the two processes are similar, with the major differences in the raw material and its pre-treatment, alcohol, catalyst costs (including the catalyst preparation and regeneration), and the separation steps in biodiesel/by-products purification. The investment costs for process A include (1) the powder mill and furnace for the catalyst preparation section, (2) solid-liquid filter, dryer and furnace for the catalyst regeneration section, and (3) washing vessel, centrifuge and membrane filter for the pre-treatment of LTW, while its yearly pro- duction costs are divided into the expenditures of (1) LTW as the raw material, (2) WCS as the catalyst material, (3) ethanol as the alcohol source, (4) energy required to purify LTW, prepare and regenerate the catalyst, and (5) utility, which includes process water for the feedstock pre-treatment. On the other hand, the traditional process B requires (1) mixing vessel and centrifuge for biodiesel purification, and (2) mixing vessel for the neutralization of acidic glycerine (by-product). The an- nual operational costs of the classic transesterification process B are represented by five major expenses, namely RPO as feedstock, methanol as acyl acceptor (12 wt% of RPO1), sodium methylate 30 % solution in methanol (1.67 wt% of RPO1) as the homogenous catalyst, hydrochloric acid (1.2 wt% of RPO1) and liquid caustic soda (0.12 wt% of RPO1) for biodiesel washing and glycerine purification, as well as the energy needed for the separation. The other expenditures are considered neg- ligible since aside from the mentioned operational steps, both of the processes are executed in a similar manner. Meanwhile, the annual gross revenues for the biodiesel production consists of the sum of credit obtained by selling of main product (bio- diesel) and by-products (glycerol and fatty matter). The yield of main and side products in this proposed technique was assumed to be the same as that of the traditional one, leading to a negligible value of the product revenues. The summary of the investment costs of the two processes is presented in Table 4, while the corresponding operating costs are given in Table 5. As presented in Table 4, having the same capacity of 9.44 tons/h, the investment cost for process A is higher by 2.9 folds as compared to process B, since process A requires more processing equipment. However, the economic analysis of their operating costs (Table 5) showed that the total processing cost of process A takes only 17.4 % of that for process B, even though the energy consumption of process A is more than 4 times higher than its corresponding value for process B. Using waste-originated feedstock and catalyst played the significant role in reducing the operating expenditures. This result is in agreement with the studies reported by Santosa

2**et al**. (2019) **and** Soufi **et al**. (2017), which **stated that the**

cost of feedstock and supporting chemicals gen- erally pose as the major operating expenditures [18,65]. Therefore, the 1 The required composition of supporting materials in process B were ob- tained from a local biodiesel manufacturer in Indonesia. Table 3 Fuel

1**Properties of LTW-based biodiesel. Properties Methods** Unit **ASTM D6751**

-19 LTW-based biodiesel Kinematic viscosity (at 40 °C) Cetane number Acid value Cloud point Flash point Heating value

3**ASTM D445 ASTM** D613 **ASTM** D664 **ASTM** D2500 **ASTM**

D93 ASTM D240 mm2 s−1 – mg KOH/g °C °C MJ kg−1 1.9 – 6.0 47 min 0.50 max Location and season dependent 93 min – 4.5 51.6 0.21 10.1 171 44.71 Table 4 The comparison of investment cost between process A and B. Units Cost (US$)* Power (kWh) Process A Pre-treatment of LTW Washing vessel Centrifuge Membrane filter Catalyst preparation Powder mill Furnace Catalyst regeneration Solid liquid filter Dryer Furnace Total investment cost Process B Biodiesel purification Washing vessel Centrifuge Glycerine purification Neutralizing tank Total investment cost 24,590 110,057 70,127 49,946 51,077 44,874 49,583 51,077 451,331 24,590 110,057 22,375 157,022 35 33 22 250 35 38 4 35 452 35 33 35 103 * Prices were obtained from the local machinery supplier. Table 5 Economic analysis of operational costs for process A and B. Input Required amounts Unit price Cost (US$) (US$) Process A LTW (ton) Water (ton) WCS (ton)a Ethanol (ton) Energy cost (kWh) Total operating costb Process Bc RPO (ton) Sodium methylate 30 % solution in methanol (ton) Methanol (ton) Hydrochloric acid (ton) Liquid caustic soda (ton) Energy cost (kWh) Total operating costb 100,000 300,000 2,242.2 25,194.7 3,579,840 74,740 1,248.16 8,968.8 896.88 89.69 815,760 – 2.99 900 312 0.11 750 2500 540 260 470 0.11 – 897,000 2,017,980 7,860,746 393,782.4 11,169,508.4 56,055,000 3,120,395 4,843,152 233,188.8 42,154.3 89,733.6 64,383,623.7 a Assuming that there is no recycled WCS-based CaO used in the process, the required amount of WCS was directly expressed by the total WCS needed to convert the main product. b The total operating costs of process A and B were calculated on the same basis of duration (continuous production for 330 days) and production capacity (74,740 tons of biodiesel). c The required amount of raw/supporting materials in process B and its prices were obtained from a local biodiesel manufacturer in Indonesia. utilization of LTW and WCS as the materials to produce biodiesel could be significant support for the zero-waste act and the feasible solution to the energy challenge. 4. Conclusions The WCS-based catalyst was successfully used as a heterogeneous catalyst for biodiesel preparation from LTW. The maximum FAEE yield was 93.4 wt%, obtained at the temperature of 60 °C and 4 h reaction time using catalyst loading of 3 wt% and ethanol to LTW molar ratio of 6:1, with the purity of 97.8 wt%. The study proved that the WCS-based catalyst has comparable activity to the other heterogeneous catalysts. The viability study concluded that the positive use of WCS and LTW in the energy-related sector provided a major benefit for the zero-waste act and is a potential key in solving the energy problems.

3**CRediT authorship contribution statement** Maria Yuliana: **Conceptualization, Methodology, Software**, Visualization, Investigation, **Writing - original draft**

. Shella Permatasari Santoso: Visualization, Investigation, Writing - original draft.

3**Felycia Edi Soetaredjo: Resources**, Data curation. **Suryadi Ismadji: Resources**

, Writing - review & editing, Supervision. Artik Elisa Angkawijaya: Software, Data curation. Wenny Irawaty:

2**Software, Validation. Yi-Hsu Ju: Writing - review & editing**, Supervision. **Phuong Lan Tran-Nguyen: Writing - review & editing**

.

2**Sandy Budi Hartono: Software, Validation, Writing - review & editing**

, Supervision.

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