



Efficient chemo-catalytic transformation of spent coffee grounds into lactic acid using erbium triflate

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ABSTRACT

Developing efficient conversion technologies for waste valorization is crucial in the context of a sustainable bioeconomy. The massive generation of spent coffee grounds (SCG) has become an environmental and social concern. Although the majority of SCG is underutilized, it exhibits the potential to be converted into value-added products. Herein, we report the effective production of lactic acid (LA), a precursor of polylactic acid, from SCG through catalytic upcycling using an erbium triflate catalyst. The reaction conditions, including time, temperature, and catalyst loading, were optimized to maximize the LA yield. The proposed strategy converted SCG into LA with a high yield (70 mol%) within 1 h. The morphology and chemical structure of SCG were investigated. Additionally, a techno-economic analysis (TEA) was conducted, which demonstrated that an economically viable catalytic upcycling strategy can enhance the efficiency of the waste valorization process.

1. Introduction

A sustainable circular bioeconomy can promote waste valorization through maximum utilization of bioresources, minimization of waste generation, and production of renewable energy that can replace fossil fuels [1,2]. Coffee is one of the world's most consumed beverages, carrying significant environmental concerns in the form of spent coffee grounds (SCG). Because SCG has no commercial value, most of them are currently discarded as solid waste. However, some studies reported that SCG can be used as feedstocks for producing fertilizer, biodiesel, and bioactive compounds [2]. Considering that coffee consumption is expected to increase, followed by the generation of massive waste streams, the valorization of SCG is important to develop a sustainable and circular economy. Despite the advances in waste valorization, developing practical and greener approaches is still an open issue in the circular economy context.

Lactic acid (LA), a precursor of polylactic acid (PLA), has gained significant attention because of the high demand for biodegradable PLA [3]. The main approaches of LA production involve chemical (acid/alkali) or thermal pretreatment of sugar-based biomass to release fermentable sugars, followed by biological conversion to the final product. However, conventional technologies require a multi-step process, the use of expensive enzymes, and a relatively long fermentation time (36 – 216 h) [4,5]. As an alternative, a chemo-catalytic conversion strategy has been proposed to produce LA from biomass and waste feedstocks. Chemical conversion, typically as a single process, has the inherent advantage of manufacturing compounds in a short time, which can minimize waste generation and does not require a separate pretreatment step [6,7]. In this regard, establishing a reliable chemical conversion process is of great importance in modern biorefinery.

Lanthanide triflate is a promising and highly effective catalyst that is stable in water, easy to recover for reuse, and can be applied in

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biorefineries to achieve a sustainable circular bioeconomy [8,9]. Chemo-catalytic conversion strategy using lanthanum triflate can effectively produce LA faster than a fermentation-based process [10]. In the previous study, several lanthanide triflate catalysts were tested to produce LA from various feedstocks, including cellulose [9], hemicellulose [10], and waste corrugated cardboard [8]. Of the tested lanthanide triflates, Er(OTf)₃ was the best catalyst to transform carbohydrates to LA. Obviously, the chemo-catalytic conversion process using lanthanide triflate is feedstock-agnostic to produce LA with decent performance. It is clear that developing scalable conversion technologies can reduce the environmental impact of the chemical industry [11].

Techno-economic analysis (TEA), as a critical analytical tool, can help determine the cost-effectiveness of a waste biorefinery and the economic feasibility of the process [12]. There have been several TEA studies on LA production from waste feedstocks. For example, LA can be produced from waste corrugated cardboard at a minimum selling price (MSP) of \$790 to \$10,370 per metric tonne, depending on the lanthanide triflate catalysts [8]. These estimates are comparable to literature estimates for LA prices that range from \$840 to \$3800 per metric tonne using various biomass feedstock and technology combinations [13–15]. Er(OTf)₃ has been identified as a cost-effective catalyst with the potential to produce LA at \$790 per metric tonne MSP. This TEA investigates the economic performance of Er(OTf)₃ for converting rarely reported SCG to LA under various reaction conditions.

The present study attempted to upcycle SCG for LA production through chemo-catalytic conversion using Er(OTf)₃, one of the best performing catalysts to produce LA from carbohydrates. The reaction conditions were optimized to maximize LA yields. In addition to LA, fatty acids released from the conversion reaction were analyzed, ensuring the full utilization of SCG. Furthermore, the morphological and structural changes after catalytic conversion of SCG were examined to assess the LA production process. Finally, TEA was carried out to evaluate the economic feasibility of the proposed chemo-catalytic approach. The findings of this study are expected to provide significant insights toward the development of a waste-free and sustainable bioeconomy.

2. Materials and methods

2.1. Materials

SCG samples used in this work were locally collected (Seoul, Republic of Korea). After collection, the SCG samples were immediately dried in an oven at 105 °C for 24 h. Table 1 shows the content of SCG components, which was determined by the National Renewable Energy Laboratory protocol [16]. Chemicals, including Er(OTf)₃, linoleic acid, palmitic acid, glucose, and xylose were purchased from Sigma-Aldrich (St. Louis, MO, USA). Analytical grade DL-lactic acid was purchased from Tokyo Chemical Industry Co, Ltd (Tokyo, Japan).

2.2. Chemo-catalytic transformation of SCG into LA

Conversion of SCG into LA was conducted using a 50 mL high pressure reactor (Parr Instrument Company, Moline, IL, USA). In a

typical reaction, 200 mg of SCG and 0.05 – 0.20 mmol of Er(OTf)₃ were placed in the reactor, and 30 mL of deionized water was added. After pressurizing to 500 psi with N₂, the reaction was carried out at desired reaction temperatures, ranging from 200° to 275 °C for varying reaction times (30 – 90 min). Following the reaction, the reactor was immediately removed from the heater and cooled in an ice bath. The resulting mixture was filtered using a syringe filter to remove any residual solids and directly injected to HPLC system to analyze reaction products.

2.3. Analytical methods

LA produced after SCG catalytic conversion was quantitatively analyzed using a YL9100 HPLC (Young-Lin, Seoul, Republic of Korea) equipped with a refractive index detector. The mobile phase was 5 mM H₂SO₄ at a constant flow rate of 0.6 mL min⁻¹. A Bio-Rad Aminex HPX-87 H column (Bio-Rad Laboratories, Hercules, CA, USA) was used for analysis. Analytical grade LA standards were used for peak identification and to prepare calibration curves for quantification. The temperature of the column compartment was maintained at 60 °C. The LA yield was calculated by the Eq. (1).

$$LA\ yields(\text{mol}\%) = \frac{\text{moles of LA}}{2\ \text{moles of C6 sugars} + \text{moles of C5 sugars in SCG}} \times 100 \quad (1)$$

After catalytic conversion, free fatty acids were extracted by adding 10 mL of ethyl acetate to the final solution. The extracted fatty acids were analyzed using Agilent 7820 A gas chromatography equipped with an Agilent 5975 mass spectrometry detector (Agilent Technologies Inc., Santa Clara, CA, USA). An Agilent HP-5 MS UI column (30 m × 0.25 mm × 0.25 μm) was used for product separation. Injection temperature was maintained at 250 °C. Oven temperature was preset to hold at 70 °C for 5 min, ramp to 250 °C at 10 °C min⁻¹, and then hold for 5 min

The structural changes of SCG before and after the catalytic process were determined using a Nicolet iS10 Fourier Transform Infrared Spectrometer (FTIR, Thermo Fisher Scientific, Waltham, MA, USA). In this work, the FTIR spectra collected were within the data range 500 – 4000 cm⁻¹ at a resolution of 4 cm⁻¹.

The morphological changes of the SCG before and after the catalytic process was examined using an Inspect F scanning electron microscopy (SEM, Thermo Fisher Scientific, Waltham, MA, USA). After reaction, the residual solids were washed with an excessive amount of DI water to remove any impurities and dried in an oven at 105 °C for 24 h. Before acquiring SEM images, a thin layer of platinum was coated onto the dried samples using an E-1045 vacuum-ion sputter coater (Hitachi, Ibaraki, Japan). SEM images were recorded with an accelerating voltage of 5.0 – 10.0 kV.

2.4. Techno-economic analysis

TEA employs an Aspen Plus TM model to simulate facility unit operations and gather material and energy balances. Capital and operating costs were gathered from Aspen Process Economic Analyzer TM. Installation factors are based on Peters and Timmerhause [17]. Labor costs, utility prices, and other miscellaneous assumptions are based on national laboratory design reports [18,19]. Details of this model and its assumptions are described in our previous study [8]. The common assumptions for this analysis include 1) a commercial scale capacity of 200 metric tons per day input of SCG, 2) a 20-year project lifetime, 3) annual operating 7884 h per year, 4) an income tax rate of 39%, and 5) a target internal rate of return (IRR) of 10%. We calculate the MSP with a discounted cash flow rate of return analysis that achieves a net present value of \$0 at the desired IRR.

2.5. Response surface methodology

A statistical full factorial design was used to examine the effect of two

Table 1
Chemical compositions of the untreated SCG.

Components	Compositions (wt%)
Carbohydrates	50.35
Cellulose	9.46
Hemicellulose	40.89
Lignin	25.34
Acid-insoluble lignin	21.26
Acid-soluble lignin	4.08
Extractives	13.72
Ash	1.77
Total (%)	91.18

factors (i.e., temperature and time). Each factor has three levels with two replicates (Table 2). The two factors, time and temperature were represented by the variables, X_1 and X_2 , respectively. In this work, the response was LA yield. The second-order polynomial, as shown in Eq. (2), was calculated to estimate the response of the dependent variables (i.e., Y) based on obtained experimental results, where a_n represents coefficients.

$$Y = a_0 + a_1X_1 + a_2X_2 + a_3X_1X_2 + a_4X_1^2 + a_5X_2^2 \quad (2)$$

All the statistical analysis was performed using RStudio software (2022.12.0). Analysis of variance was utilized to figure out the statistical significance of each parameter. The response surface was drawn to understand the interactions between variables.

3. Results and discussion

3.1. Chemo-catalytic conversion of spent coffee grounds to lactic acid

Compositional analysis of SCG indicates that carbohydrates constitute the predominant component, comprising 50 wt% (Table 1). This is followed by lignin, extractives and ash. It is worth noting that SCG may also contain a minimal quantity of proteins. The catalytic conversion of SCG was performed using $\text{Er}(\text{OTf})_3$ catalyst at varying temperatures (200 – 275 °C) and reaction times (30 – 90 min) to find the optimal conditions to produce LA. Previously, several lanthanide triflate catalysts, including $\text{La}(\text{OTf})_3$, $\text{Nd}(\text{OTf})_3$, $\text{Yb}(\text{OTf})_3$, and $\text{Er}(\text{OTf})_3$, were examined to convert waste corrugated cardboard, and $\text{Er}(\text{OTf})_3$ was the best catalyst to convert sugars into LA [8]. $\text{Er}(\text{OTf})_3$ catalyst with a small ionic radius as water-tolerant Lewis acids among the lanthanide triflates was reported to exhibit higher catalytic activity by strongly coordinating with hydroxyl groups of sugars [20]. The present study, therefore, focuses on $\text{Er}(\text{OTf})_3$ for producing LA from SCG.

Fig. 1 shows LA yields from SCG samples after catalytic conversion. As shown, the LA yield increased with increasing temperature from 200 to 250 °C at each reaction time, and a high LA yield (62 mol%) was obtained at 250 °C for 60 min. It was reported that the Brønsted acidity of the catalyst accelerates the hydrolysis of sugar at high temperatures and effectively converts it into LA [7]. However, at 275 °C, the yield of LA noticeably decreased at all the reaction times tested in the present study. The decrease in LA yields is attributed to the thermal decomposition of the products at higher temperatures. The effect of reaction time on LA yields was marginal at 200 – 250 °C, whereas the LA release significantly decreased when the reaction time increased to 90 min at 275 °C. As discussed above, thermal degradation of the produced LA possibly occurs at 275 °C, which is facilitated at an extended reaction time.

The empirical model for LA yield (mol%), developed from the statistical analysis is shown in coded form in Eq. (3). The R^2 value of the model is 0.789, indicating a good fit of the data to the models.

$$Y = 61.926 - 0.389X_1 - 1.550X_2 - 2.529X_1X_2 - 0.494X_1^2 - 4.797X_2^2 \quad (3)$$

A t -test was performed for each parameter to determine the significance of each coefficient developed and the results are presented in Table 3. As indicated, temperature, interaction term of time and temperature, and a second-order term of temperature are found to be statistically significant since p -values are less than 0.05. Furthermore, the

Table 2
Full factorial design for catalytic conversion experiments.

	Variables		Coded levels	
	Time (min, X_1)	Temperature (°C, X_2)	X_1	X_2
Low level	30	225	-1	-1
Medium level	60	250	0	0
High level	90	275	+1	+1

analysis of variance of the model shows the high F -values and low p -values, indicating that these models are statistically significant, and there are no significant lack of fit (Table 4).

Fig. 2 illustrates the response surface of LA yields as a function of the independent variables, reaction time and temperature. Apparently, the reaction temperature is more influential than reaction time. The analysis indicates that the stationary point of the fitted surface is at 61.81 min and 245.56 °C, which is well within the experimental region.

In addition to reaction temperature and time, the effect of catalyst loading on the production of LA was explored. It was hypothesized that the higher catalyst loading could produce more LA. To test the hypothesis, the catalyst loading was further increased to 0.1 and 0.2 mmol, and the same conversion reaction was performed. Fig. 3 shows LA yields after chemo-catalytic reaction with different catalyst loadings. Obviously, the yield of LA significantly increased from 62 to 70 mol% with an increasing catalyst loading (0.1 and 0.2 mmol). This result suggests that a higher amount of catalyst provided more acidic sites during the cascade reaction for effective sugar hydrolysis [21], resulting in high conversion into LA. Although the economic viability of the proposed process with different catalyst loadings has to be assessed (see Section 3.2), the catalytic conversion strategy can effectively transform SCG into a value-added product.

There have been enormous efforts to recover carbohydrates and sugars in SCG because they can be renewable sources for producing fuels and chemicals. Physical, chemical, and biological approaches have been implemented to obtain sugars from SCG [22]. However, most of these processes have certain limitations. For example, pretreatment typically requires the use of toxic chemicals, followed by washing to remove undesirable chemicals, which generates a large quantity of wastewater and results in high operating costs [6]. Therefore, it is time to come up with an alternative, greener, and simpler process for the efficient conversion of carbohydrates, even without pretreatment steps. In this respect, the proposed chemo-catalytic conversion that 1) directly transforms carbohydrates and sugars to a value-added building block, 2) uses water as a solvent, and 3) can be operated with fewer unit operations would be a good strategy for waste valorization, promoting a circular economy.

It is also noteworthy that a significant amount of fatty acids was produced from the catalytic conversion of SCG. The extraction of fatty acids and biodiesel production from SCG have been widely studied due to their high lipid contents. Since lipids extraction is typically conducted using organic solvents at elevated temperatures [23], the observation of fatty acids from the catalytic conversion in an aqueous media is unexpected. The release of fatty acids from the SCG is possibly due to the catalytic effect and relatively high temperatures used in the present work [24]. To analyze the fatty acids, ethyl acetate was added to the resulting solution, and the organic layer for amount of fatty acids was quantified by GC. Fig. 4 shows the representative GC chromatogram and the amount of fatty acids quantified. The two primary fatty acids present are palmitic acid and linoleic acid. These results are in good agreement with the previous reports for the extraction of lipids from SCG [25–27]. The amount of palmitic acid and linoleic acid was 3.1 and 4.9 wt%. Although the extraction conditions of lipids are not optimized in the present work, the proposed strategy exhibits the potential to generate a secondary coproduct stream in addition to LA production from SCG. Besides fatty acids, several peaks assigned to lights oxygenates, possibly derived from carbohydrates and lignin, were observed; however, the amount of products were substantially marginal.

Understanding the structural changes of SCG after catalytic reactions is of great importance in identifying the potential application areas [28]. FTIR analysis revealed that SCG has absorption bands of polysaccharides (Fig. 5). The broad peak of O–H functional groups (3300 cm^{-1}) and C–O stretching (1030 cm^{-1}) are related to the arabinogalactans present in SCG [29,30]. The signal intensity of these peaks diminished from the residual solid obtained after the conversion reaction. These results suggest that the carbohydrates present in SCG are effectively hydrolyzed

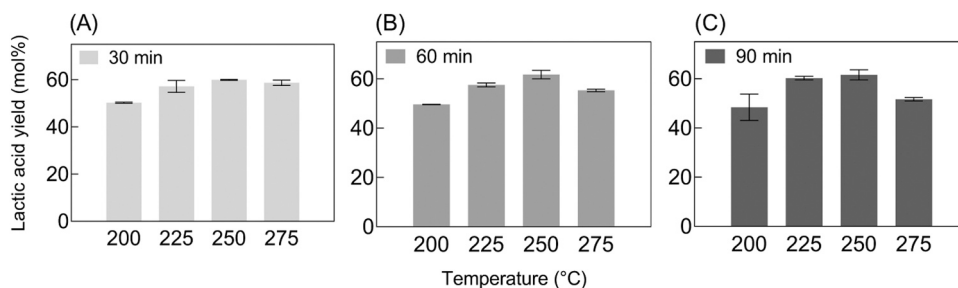


Fig. 1. LA yields after catalytic conversion of SCG with $\text{Er}(\text{OTf})_3$ at varying reaction temperatures for (A) 30 min, (B) 60 min, and (C) 90 min (Catalyst loading: 0.05 mmol).

Table 3

Parameter estimates of intercepts, individual parameters and interaction.

Term	Estimate	Std Error	t value	p > t
Intercept	61.92611	1.04036	59.5237	< 0.0001 ***
X_1	-0.38917	0.56983	-0.6830	0.5076
X_2	-1.55000	0.56983	-2.7201	0.0186 *
$X_1 \times X_2$	-2.52875	0.69789	-3.6234	0.0035 **
X_1^2	-0.49417	0.98697	-0.5007	0.6256
X_2^2	-4.79667	0.98697	-4.8600	0.0004 ***

Table 4

Analysis of variance for the model based on experimental data.

	DF	SS	MS	F value	P (>F)
FO (X_1, X_2)	2	30.647	15.324	3.9327	0.048584
TWI (X_1, X_2)	1	51.157	51.157	13.1290	0.003493
PQ (X_1, X_2)	2	93.009	46.504	11.9350	0.001402
Residuals	12	46.758	3.896	-	-
Lack of fit	3	18.007	6.002	1.8789	0.203580
Pure error	9	28.751	3.195	-	-

DF: Degrees of freedom, SS: Sum of square, MS: Mean square

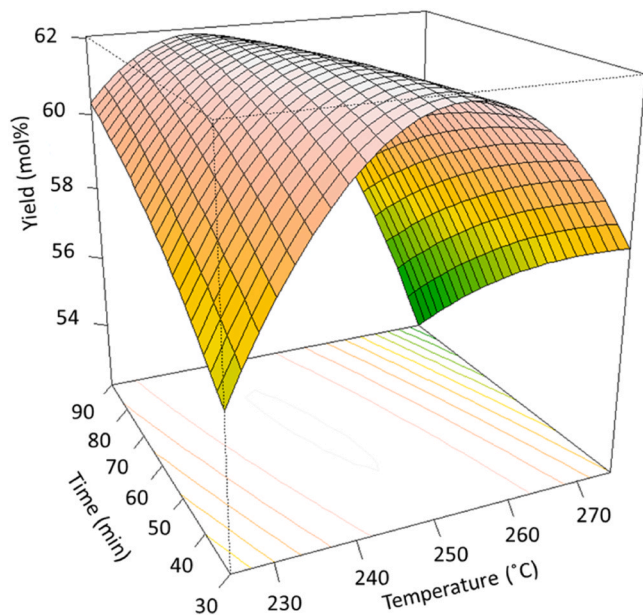


Fig. 2. Surface contour plot of LA yields vs. reaction time and temperature. Stationary point of response surface: $X_1 = 0.06031$, $X_2 = -0.1775$. Stationary point in original units: time = 61.81 min, temperature = 245.56 °C.

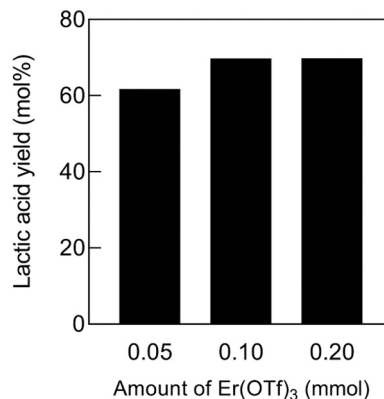


Fig. 3. Effect of catalyst loading on LA yield. (Reaction conditions: 250 °C and 60 min).

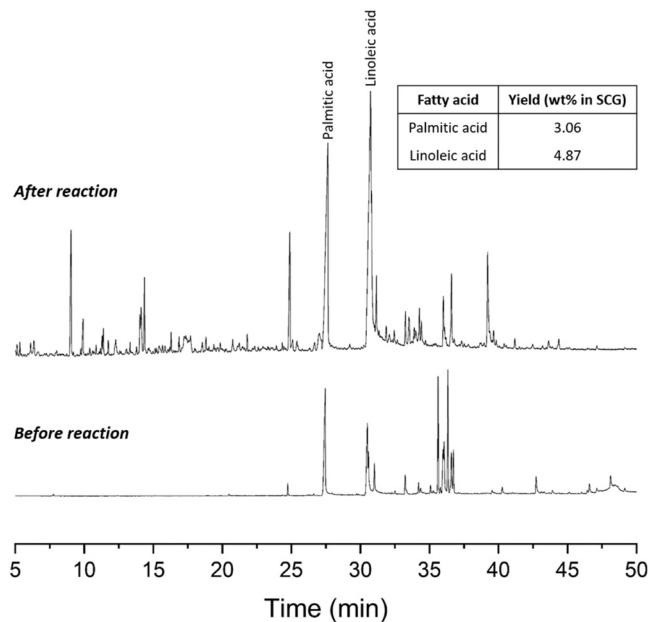


Fig. 4. Gas chromatogram of products in the organic layer and the amounts of fatty acids obtained from catalytic conversion of SCG (Reaction conditions: 0.1 mmol of catalyst loading, 250 °C, and 60 min).

and converted to LA by the $\text{Er}(\text{OTf})_3$ mediated reaction [31]. C-H stretch vibrations (2850 and 2920 cm^{-1}) and peaks with bands of $\text{C}=\text{O}$ (1640 and 1740 cm^{-1}) indicate the presence of residual caffeine [30]. The major components, including caffeine, phenolic compounds, and carotenoids, are still present in SCG after the catalytic process. These

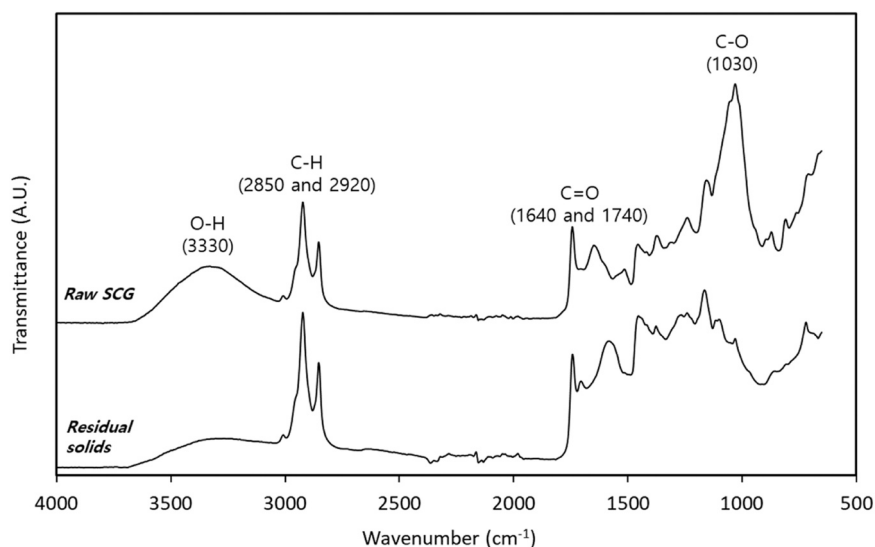


Fig. 5. FTIR spectra of SCG before and after chemo-catalytic conversion.

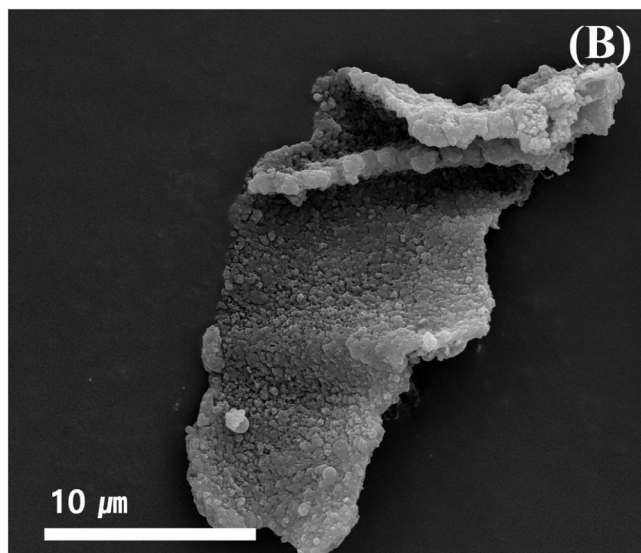
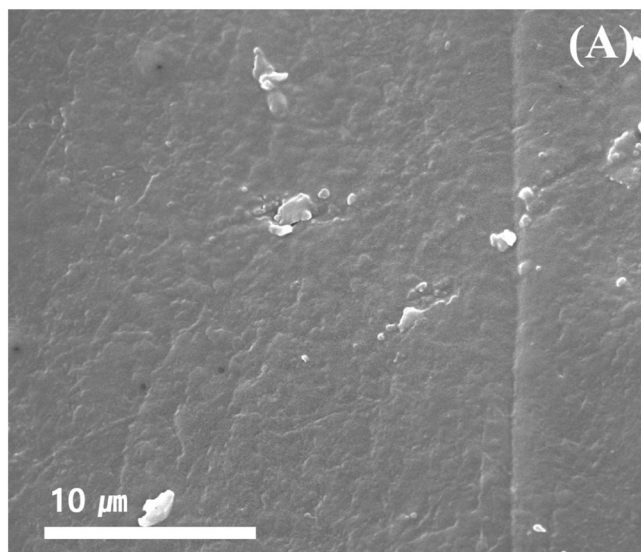


Fig. 6. SEM image showing the surfaces of the SCG before (A) and after (B) chemo-catalytic conversion.

valuable chemicals can be further utilized through appropriate chemical and biological processes [22].

Morphological changes of SCG were analyzed by SEM (Fig. 6). The surface of the untreated SCG was smooth and non-porous. After the catalytic process, however, the residual solid showed atrophic and wrinkled shapes, possibly with increased surface area. Physico-structural changes in the biomass morphology suggest improved extraction efficiency of carbohydrates in SCG for LA production [32]. Residual SCG can be further utilized as biochar for environmental remediation, such as bio-adsorbents, carbon storage medium, and soil amendment [22].

3.2. Techno-economic analysis

For TEA, we conducted a scenario analysis based on the LA yield, reactor residence time, and reactor temperature. The process model includes SCG pretreatment, chemo-catalytic conversion, esterification, hydrolysis, and distillation [8]. A simplified process flow diagram was proposed (Fig. S2). Capital and operating costs and the MSP were calculated for each scenario, and Fig. 7 shows the results of the scenario analysis. As shown, the LA MSP ranges between \$728 and \$1107 per metric tonne, depending on reaction conditions. The MSP decreased with temperature except for the 90 min, 275 °C scenario due to the increase in LA yields. The MSP increases with residence time due to higher reactor and catalyst costs. The lowest cost of \$728 per metric tonne was achieved at 30 min residence time and 250 °C. The MSP for 30-min residence time and 275 °C is statistically similar at \$744 per metric tonne.

As discussed earlier, higher LA yields were observed when the catalyst loading increased from 0.05 to 0.10 mmol. We compared the impact of the increased catalyst loading in the reactor from 0.05 mmol to 0.10 mmol, and the results are shown in Table 5. It is clear that increased catalyst loading results in greater LA production and revenue. However, the MSP is higher with a high catalyst loading (\$782.7 vs. \$805.5) because of the increased capital cost, suggesting that determining the right amount of catalyst is crucial to maximize profit.

This analysis provides an economic basis for comparing the operating conditions for converting SCG to LA using an $\text{Er}(\text{OTf})_3$ catalyst. Catalyst lifetime and byproduct credits are two factors not considered in this analysis that could change the conclusions. Catalyst replacement costs could increase if higher temperatures and longer residence time decrease the catalyst lifetime. Additionally, it should be pointed out that byproduct credits could compensate for scenarios with lower LA yields.

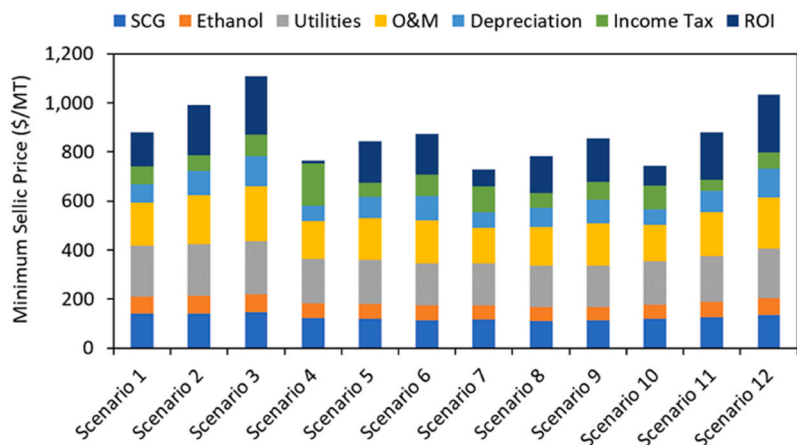


Fig. 7. Minimum selling price of LA depending on each scenario.

Scenario	Conditions	MSP (\$/MT)
1	30 min, 200 °C	880.6
2	60 min, 200 °C	990.4
3	90 min, 200 °C	1107.5
4	30 min, 225 °C	765.8
5	60 min, 225 °C	843.7
6	90 min, 225 °C	874.5
7	30 min, 250 °C	728.5
8	60 min, 250 °C	782.7
9	90 min, 250 °C	854.3
10	30 min, 275 °C	744.7
11	60 min, 275 °C	879.8
12	90 min, 275 °C	1032.0

Table 5

Summary of TEA based on two catalyst loading scenarios.

Scenario	Low catalyst loading (0.05 mmol)	High catalyst loading (0.10 mmol)
Lactic acid production (kg/year)	11,930	13,580
Annualized costs (\$/MT)		
Income tax	84.83	89.65
Ethanol	56.02	49.21
Depreciation	79.13	98.85
SCG	111.65	98.09
O&M	156.56	162.78
ROI	126.78	159.49
Utilities	167.77	147.40
Capital cost (\$MM)	18.90	26.80
MSP (\$/MT)	782.74	805.47

Future work could investigate the impacts of these tradeoffs to improve the economic characterization of this process.

Finally, this work shows that a valuable building block can be produced from the chemo-catalytic conversion of SCG. Although follow-up studies, including catalyst recycling and product separation, are needed to make the suggested process more economically feasible, the chemo-catalytic approach could facilitate the development of a circular economy.

4. Conclusions

This work demonstrates the extended potential of a chemo-catalytic conversion strategy to produce LA from underutilized SCG. The use of water as a solvent and highly efficient Er(OTf)₃ catalyst provides an opportunity to develop sustainable upcycling technology. The highest LA yield of 70 mol% was achieved within 60 min under the optimal reaction conditions. In addition, fatty acids were readily obtained following the catalytic conversion of SCG. TEA indicated a substantial economic benefit of catalytic conversion of SCG into LA. Although more in-depth studies, including catalyst recycling, product recovery, and purification will be necessary to validate the economic viability of our strategy, these results demonstrate that the catalytic upcycling of waste into valuable chemicals could facilitate the development of a sustainable bioeconomy.

CRedit authorship contribution statement

Geon-Soo Ha: Conceptualization, Methodology, Validation, Writing – original draft, **Hanseob Jeong:** Conceptualization, Methodology,

Validation, **Ho Seung Song:** Methodology, Validation, **Da Hae Oh:** Methodology, Validation, **Mark Mba-Wright:** Methodology, Validation, Writing – original draft, **Jeong-Myeong Ha:** Methodology, Validation, **Chun-Jae Yoo:** Methodology, Validation, **Jae-Wook Choi:** Methodology, Validation, **Chang Soo Kim:** Methodology, Validation, **Byong-Hun Jeon:** Methodology, Validation, **Kwang Ho Kim:** Conceptualization, Supervision, Validation, Writing – review & editing.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jece.2023.110682](https://doi.org/10.1016/j.jece.2023.110682).

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