

Conference paper

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Conversion of glucose into lactic acid using silica-supported zinc oxide as solid acid catalyst

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Abstract: Zinc oxide (ZnO) has been proven to be highly effective in converting biomass into fine chemicals. It possesses several limitations, such as leaching in hydrothermal reactions and difficulty with regard to its recovery. Supporting ZnO on silica improves its recovery, stability and recyclability. In this study, we produced silica-supported ZnO by incipient wetness impregnation (IWI) method for the conversion of glucose into lactic acid. The presence of the ZnO provided active sites for isomerization to occur. The highest yield of lactic acid was 39.2% at 180 °C for 60 min. Prolonged reaction time and higher reaction temperature promoted further degradation of lactic acid into acetic acid. The yield of lactic acid decreased after the first cycle and decreased slightly for the nine consecutive cycles.

Keywords: lactic acid; POLYCHAR-25; recyclability; supported catalyst; zinc oxide.

Introduction

Global warming and depletion of fossil fuel reserves have driven us to seek alternative energy resources with renewable carbon feedstocks, such as lignocellulosic resources [1–3]. Lignocellulosic biomass consists mainly of lignin, hemicellulose and cellulose, which are useful and can be converted into various valuable chemicals, including fuel additives, biopolymers, resins and solvents [4, 5]. Lactic acid (LA) is one of the platform chemicals obtained by the conversion of sugar precursors. Moreover, LA can be used in food preservation, cosmetics and polymers. Particularly, LA can be polymerized into poly(lactic acid), which is a widely used biodegradable polymer that can reduce the reliability of petroleum-based polymers [6, 7].

LA is commercially produced by fermentation method. However, this method has several disadvantages, such as high production cost, tedious production conditions, difficulties in microbe recovery and the formation of salt wastes [8, 9]. Therefore, catalytic conversion of sugars into LA using Lewis acid, Brønsted base and transition metal salts has been studied by researchers [10–12]. Yet, these homogeneous catalysts have

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several disadvantages, such as low recyclability, corrosiveness and the requirement of post-treatment of the hydrolysate [5]. Hence, heterogeneous catalysts have been attracting much attention recently [13].

Solid acid catalysts can be used as heterogeneous catalysts due to the presence of Brønsted acid sites. There are various types of solid acid catalysts, including metal oxides, polyoxometalates, zeolites and supported acids [14–17]. In the field of supported catalysts, various supports, such as silica, aluminum oxide, tin oxide (SnO_2) and carbon, have been used previously. Silica possesses several advantages, such as irreducible and inert properties, high thermal stability, large pore size and high specific surface area for catalyst impregnation [13, 18]. The advantages of a supported catalyst can minimize leaching of the catalyst [19, 20]. Moreover, supporting a nano-sized catalyst can improve the recovery of the catalyst [21].

Various metal oxides, such as zirconium (ZrO_2), SnO_2 , copper oxide (CuO), calcium oxide (CaO) and magnesium oxide (MgO), have been used in the conversion of glucose into fine chemicals [14, 22–24]. Yang et al. [25] have studied the catalytic activity of (ZrO_2) in the conversion of hemicellulose to LA. The yield of LA was 30 % and 42 % from xylan and xylose, respectively [25]. Conversion of glucose into LA and gluconic acid was studied by Onda et al. [26]. They synthesized Pt that was supported on activated carbon, and the catalyst yielded 47 % and 46 % LA and gluconic acid, respectively, from glucose [26].

In this study, we synthesized silica-supported zinc oxide ($\text{SiO}_2\text{-ZnO}$) using incipient wetness impregnation (IWI) to study its catalytic activity in the conversion of glucose into fine chemicals, particularly LA.

Experimental

Materials

D-glucose (purity $\geq 99\%$), D-fructose (purity $\geq 99\%$), zinc chloride (ZnCl_2 , purity), lactic acid (purity $\geq 99.5\%$), 5-hydroxymethylfurfural (5-HMF, purity $\geq 99.5\%$), acetic acid (purity 50 %), 25 % ammonia solution (NH_4OH), 65 % nitric acid (HNO_3) and silica gel 60 (mesh 70–230) were used for column chromatography. All of the chemicals were analytical-grade and purchased from Sigma Aldrich, whereas NH_4OH , HNO_3 and silica gel 60 were purchased from Merck.

Catalyst preparation

Silica gel 60 and ZnCl_2 were mixed at a ratio of 2:1 and dissolved in 0.5 M NH_4OH solution. The mixture was stirred for 3 h, washed with deionized water twice and dried in a freeze dryer overnight. The sample was calcinated at 500 °C for 6 h in nitrogen (N_2) atmosphere [27, 28].

Characterization

The prepared catalyst was placed on the mold and analyzed on a Bruker/D8 Advance X-ray diffractometer (XRD) with Cu K- α radiation at 0.15406 nm to determine the formation of ZnO on the silica surface. Further, the samples underwent Brunauer, Emmett and Teller (BET) analysis and analyzed on a Micromeritics ASAP 2010. The samples were degassed at 373 K for 5 h and then analyzed at 77 K. Furthermore, the morphology of the catalyst was observed under a scanning electron microscope (SEM) (Phenom ProX), and energy dispersive spectroscopy (EDS) analysis was performed.

The zinc content on the catalyst was determined on a Perkin Elmer ELAN 9000 by inductively coupled plasma-mass spectroscopy (ICP-MS). Briefly, the $\text{SiO}_2\text{-ZnO}$ catalyst was subjected to acid digestion with 20 % HNO_3 at 50 °C for 6 h. Then, the solution was diluted to 1 % HNO_3 content. For the leaching studies, the hydrolysate was filtered with a 0.22- μm syringe filter before analysis.

Catalytic performance

The catalytic reaction was performed by adding 0.1 g of catalyst into 10 mL of 1 g/L glucose solution in a boron silicate vial and heating it at 180 °C for 1 h using a microwave reactor (Anton Paar, Monowave 300, Graz, Austria). After the reaction, the catalyst was washed with deionized water and filtered with a 0.22- μm membrane filter before the second cycle of the reaction. This step was repeated several times to study the recyclability of the catalyst. After that, the hydrolysate was analyzed by high-performance liquid chromatography (HPLC) on an Agilent Technologies 1260 Infinity equipped with a Hi-Plex H column, reflective index (RI) detector and ultraviolet-visible (UV) detector at wavelength 280 nm. All of the calibration curves had $R^2 > 0.9980$. The calculation of the conversion of glucose is as follows:

$$\text{Conversion of glucose} = \frac{\text{initial mole of glucose} - \text{final mole of glucose}}{\text{mole of glucose}} \times 100\% \quad (1)$$

The calculation for the percentage yield of LA, fructose, AA and 5-HMF is shown below:

$$\% \text{ Yield of lactic acid} = \frac{\text{mole of lactic acid}}{2[\text{mole of glucose}]} \times 100\% \quad (2)$$

$$\% \text{ Yield of fructose} = \frac{\text{mole of fructose}}{\text{mole of glucose}} \times 100\% \quad (3)$$

$$\% \text{ Yield of acetic acid} = \frac{\text{mole of acetic acid}}{3[\text{mole of glucose}]} \times 100\% \quad (4)$$

$$\% \text{ Yield of 5-HMF} = \frac{\text{mole of 5-HMF}}{\text{mole of glucose}} \times 100\% \quad (5)$$

In the reaction time studies, 0.1 g of catalyst was added to 10 mL of 1 g/L glucose solution in a boron silicate vial and heated at 180 °C for 30, 45, 60, 75, 90, 120, or 150 min with a microwave reactor (Anton Paar, Monowave 300, Graz, Austria). Then, the hydrolysate was centrifuged, and the supernatant was filtered with a 0.2- μm syringe filter (Agilent). The samples were then injected into the HPLC instrument for quantitative analysis.

In the recyclability studies, the experiment was conducted based on the results obtained from the reaction time studies that had the highest yield of LA. After every cycle, the catalyst was washed with deionized water twice and filtered with a 0.22- μm nylon membrane filter before the next cycle.

Results and discussion

Characterization of SiO_2 -ZnO

The catalyst was prepared via incipient wetness impregnation (IWI)-based synthesis of ZnO on the supported silica. First, the Zn ions dissolved in water were diffused into the porous structure of silica through capillary force, where the NH_4OH was added to initiate the precipitation of $\text{Zn}(\text{OH})_2$ through aggregation. The subsequent calcination process at 500 °C led to the formation of ZnO on the SiO_2 structure [29]. Based on the ICP-MS results, the weight percentage of ZnO content in the SiO_2 support was 44.0%. The successful growth of the ZnO was confirmed via XRD analysis, as shown in Fig. 1. A broad diffraction peak at around $2\theta = 23^\circ$ implied

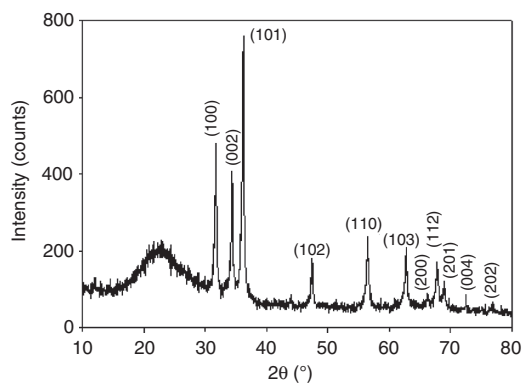


Fig. 1: XRD pattern of SiO_2 -ZnO prepared via IWI synthesis method.

that the presence of the amorphous substance was the silica [30]. The obtained XRD pattern matched the peak of JCPDS no. 36-1451, indicating the formation of a typical ZnO crystal. The surface morphology of the SiO_2 -ZnO was examined by SEM (Fig. 2). As indicated by the arrows, a flake-like structure of ZnO grew on the SiO_2 , which was further confirmed by the elemental analysis, as listed in Table 1. The EDS results showed that the Zn had the highest concentration, which contributed to the presence of ZnO. Further, the Si, O and Al contributed to the silica content, which acted as a support for the formation of ZnO. These results further support the discussion on the successful formation of ZnO. Furthermore, the effect of the growth of ZnO on the surface porosity of SiO_2 was examined using BET analysis, and the results are listed in Table 2. The decrement of BET

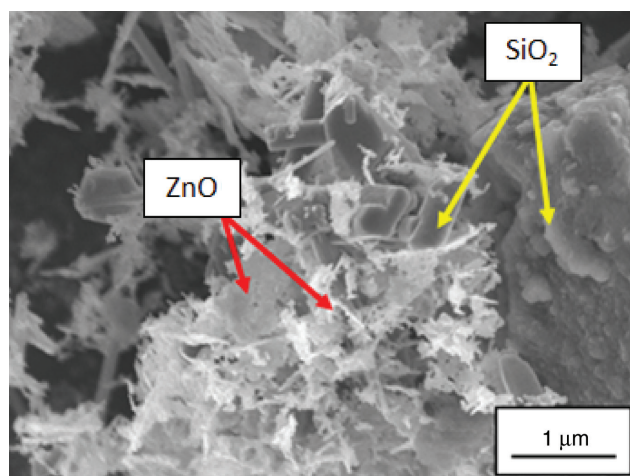


Fig. 2: Electron micrograph of SiO_2 -ZnO catalyst before catalytic reaction.

Table 1: EDS result of SiO_2 -ZnO.

Element	Atomic concentration (%)	Weight concentration (wt.%)
Zn	36.6	69.3
C	31.5	11.0
Si	11.7	9.49
O	18.4	8.53
Cl	0.84	0.86
Al	1.05	0.82

Table 2: Nitrogen absorption properties of SiO₂ and SiO₂-ZnO.

	BET surface area (m ² /g)	Total pore volume (cm ³ /g)	Pore diameter (nm)
SiO ₂	409.6	0.716	6.99
SiO ₂ -ZnO	293.2	0.554	7.56

surface area and total pore volume indicates the formation of ZnO on the SiO₂, which occupied the pores of the SiO₂ [31]. The chemical reaction for the ZnO formation is as follows:



Catalytic activity of SiO₂-ZnO

The effect of ZnO loading on SiO₂ on the catalytic conversion of glucose into LA and other compounds is demonstrated in Table 3. The blank SiO₂ yielded 1.50 % LA, which is much lower than that of the SiO₂-ZnO (24.9 %). The concentration of glucose in the hydrolysate using the SiO₂-ZnO catalyst was lower than in blank SiO₂ with the presence of ZnO, where it catalyzed the selective conversion of glucose into fructose and LA [32]. The role of the ZnO is to serve as a reservoir that is capable of inducing selective conversion of glucose into LA, which involves two stages of isomerization: (1) glucose into fructose and (2) glyceraldehyde into LA, as demonstrated in Fig. 3 [14, 25, 33]. The results also indicated a higher yield of 5-HMF for blank silica than the SiO₂-ZnO, which could be attributed to the Brønsted acid sites on the silica surface [34], whereas for SiO₂-ZnO, most of the surface was occupied by ZnO; therefore, less 5-HMF was yielded. The presence of 5-HMF could also be due to the formation of hydronium ions (H₃O⁺) in the hydrothermal reaction medium, which could promote the dehydration of glucose into 5-HMF [35].

The effect of catalyst dosage on the conversion of glucose into fine chemicals is presented in Fig. 4. The yield of LA increased from 17.5 % to 30.2 % as the catalyst that was added rose from 0.01 g to 0.2 g. The concentration of acetic acid (AA) increased slightly with increasing catalyst from 0.01 to 0.1 g. The increasing amount of ZnO made more binding sites available for catalytic activity. Therefore, more LA was yielded.

The effect of reaction temperature on the conversion of glucose into fine chemicals is depicted in Fig. 5. As the reaction temperature increased from 160 °C to 180 °C, the conversion of glucose into LA rose from 0.531 % to 3.92 % and then increased to 10.3 % at 200 °C. At 200 °C, no glucose was detected. Although higher temperature led to greater yield for LA, LA was further degraded into AA as the detected AA increased [25, 36]. Therefore, increasing temperature promotes the degradation of LA into AA.

Reaction time studies

The catalytic activity of the SiO₂-ZnO in converting glucose into different chemical compounds was further investigated by varying the reaction time from 30 to 150 min (Fig. 6). The catalytic activity in the conversion of

Table 3: Yield of conversion of glucose into fine chemicals under reaction conditions of 1 g/L of glucose and 0.1 g of catalyst in 10 mL water at a reaction temperature of 180 °C for 30 min.

Catalyst	Conversion/percentage yield (%)				
	Glucose	Fructose	Lactic acid	5-HMF	Acetic acid
SiO ₂	75.4	11.5	1.50	12.7	4.27
SiO ₂ -ZnO	88.1	18.3	24.9	3.43	3.93

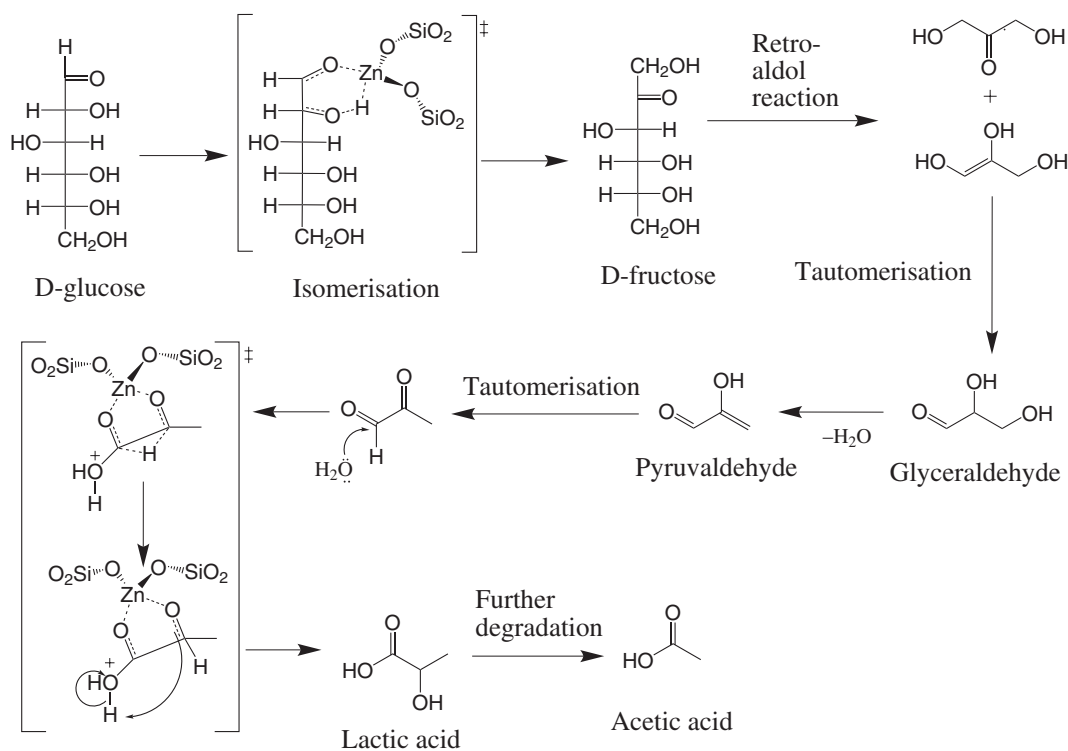


Fig. 3: Proposed reaction pathways for the conversion of glucose into LA.

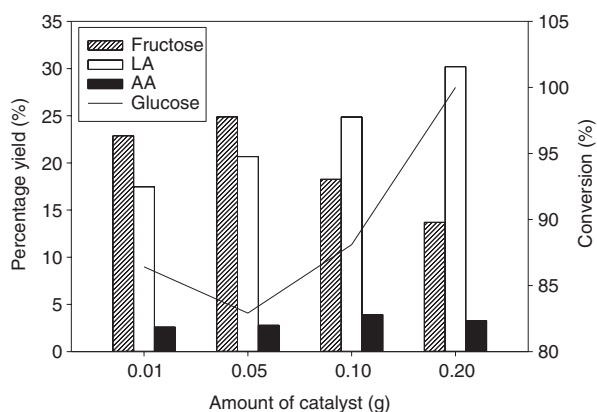


Fig. 4: Effect of various amounts of catalyst on the yield of conversion of glucose into fructose, LA and AA under reaction conditions of 1 g/L of glucose at a reaction temperature of 180 °C for 30 min.

glucose to LA increased from 30 min to 60 min and then gradually decreased. The highest yield of LA (39.2%) was obtained at 60 min. Also, the yield of LA decreased from 34.5% to 28.4% from 90 min to 150 min, which resulted in a higher AA yield due to the degradation of LA. Table 4 summarizes the catalytic performance of different previously reported heterogeneous catalysts. It can be seen that SiO₂-ZnO exhibited comparable performance in the production of LA.

Recyclability

As the catalyst was collected from the catalytic reactions of glucose into LA, the catalytic performance was further evaluated for eight consecutive cycles. The results are shown in Fig. 7. The yield of LA decreased to

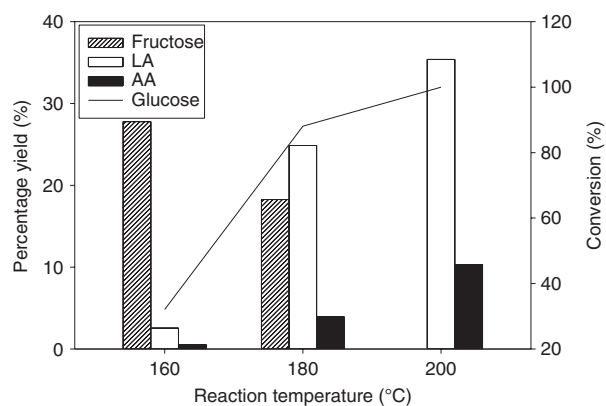


Fig. 5: Effect of reaction temperature on the yield of conversion of glucose into fructose, LA and AA under reaction conditions of 1 g/L of glucose and 0.1 g of catalyst for 30 min.

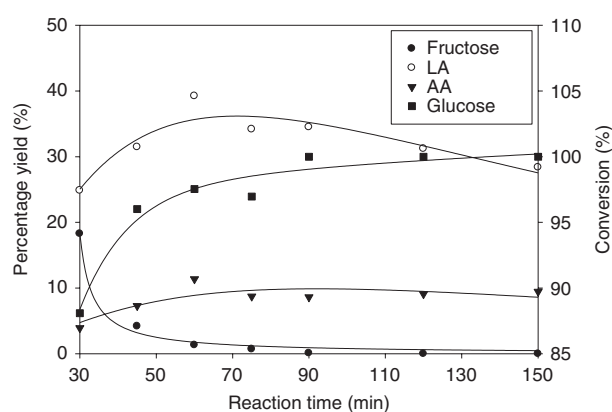


Fig. 6: Yield of conversion of glucose into fructose, LA and AA under reaction conditions of 1 g/L glucose and 0.1 g catalyst at 180 °C under various reaction times.

Table 4: Comparison studies of heterogeneous catalysts in the conversion of glucose into LA.

Source	Support	Catalyst	Yield of lactic acid (%)	Catalytic efficiency (% g ⁻¹ h ⁻¹)	Number of cycles	Ref.
Glucose	SiO ₂	ZnO	39.2	392	9	Our study
Sodium alginate	–	ZnO	0.68	1.13	–	[14]
Sodium alginate	–	CaO	12.6	21.0	5	[14]
Glucose	–	Hydrotalcite	20.3	4.23	–	[37]
Glucose	Zeolite	SnO ₂	26	8.13	–	[38]
Crystalline cellulose	–	AlW	27	1.65	3	[15]
Cellulose	Activated carbon	Zn/Ni/NaOH	42	730	–	[39]
Cellulose	Al ₂ O ₃	Er ₂ O ₃	44.1	221	3	[40]
Glucose	Zeolite	ZnO and SnO ₂	48	150	5	[32]

23.7 % as compared with the initial reaction (39.2%), which could be attributed to the leaching of loosely attached ZnO on the SiO₂ surface. This is consistent with the ICP-MS results obtained from the hydrolysate after the first and ninth cycles, during which 11.5 % and 6.1 % of the ZnO content leached out. Moreover, this can also be observed in Fig. 8, where there was agglomerated and less ZnO on the surface of SiO₂. The catalytic reactions decreased gradually after several cycles of reactions, and the yield of LA was maintained at around 20 % up to nine cycles, although the yield of LA decreased after each cycle due to the leaching of ZnO.

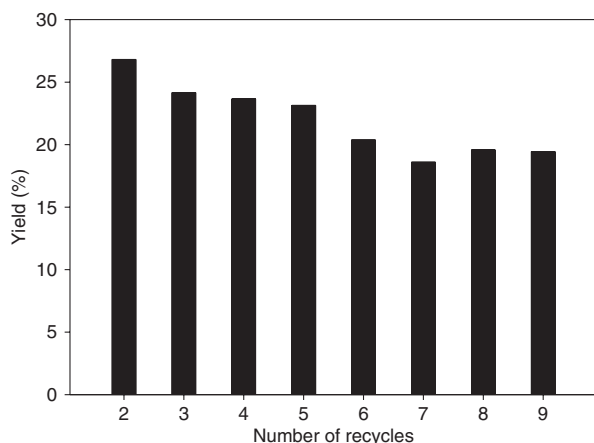


Fig. 7: Recyclability of $\text{SiO}_2\text{-ZnO}$ in the conversion of glucose into LA after the first cycle under reaction conditions of 1 g/L glucose and 0.1 g catalyst at 180 °C for 60 min.

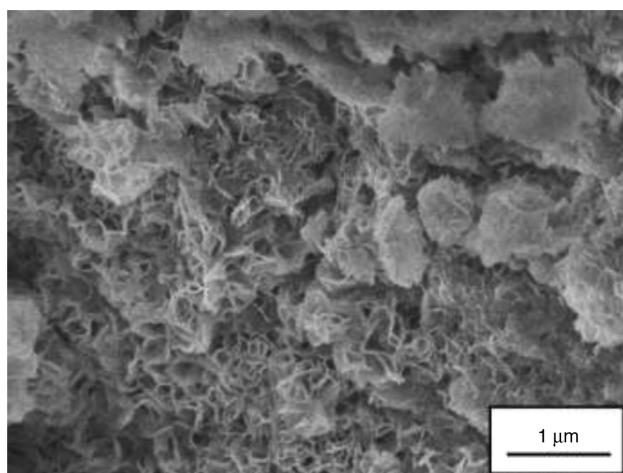


Fig. 8: SEM image of $\text{SiO}_2\text{-ZnO}$ catalyst after nine cycles of catalytic reaction of glucose to LA.

However, the results show that the $\text{SiO}_2\text{-ZnO}$ maintained its catalytic efficiency at a yield of approximately 20% of LA up to nine cycles of reactions.

Conclusion

In this study, we synthesized a $\text{SiO}_2\text{-ZnO}$ catalyst that had good catalytic activity in the conversion of glucose into LA. The highest yield of LA (39.2%) was achieved at 180 °C for 60 min. Furthermore, the recyclability studies suggested that the catalyst is stable and maintains around a 20% yield of LA up to nine cycles of reactions. A more effective system, such as a packed column, may be developed to expand the practical usability and efficiency of this catalytic system.

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