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Research Article

Rigorous Calculation of Greenhouse Gases (GHG) in Sustainable L-lactic Acid Production from Lignocellulosic Biomass based on Advanced Biorefinery Processing Technology

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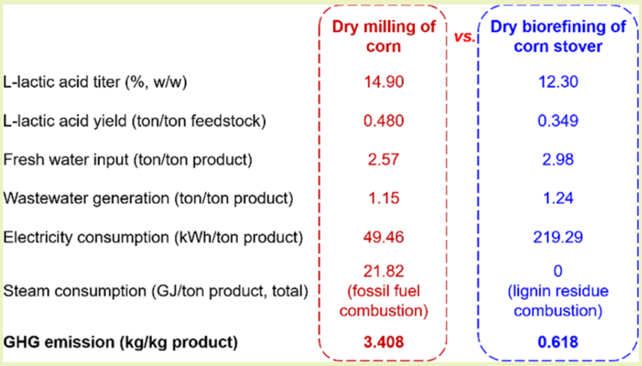
ABSTRACT: Utilizing lignocellulosic biomass to produce high-purity L-lactic acid as the monomer of poly(lactic acid) (PLA) is an important pathway for reducing greenhouse gas (GHG) emissions. This study developed an Aspen Plus simulation model and a life-cycle assessment model for producing high-purity L-lactic acid using corn stover feedstock based on thermodynamic principles and experimental data. An advanced dry biorefinery technology is selected as the process platform because of its high conversion performance, close to the dry milling of corn, as well as the significantly reduced energy consumption and wastewater generation. The GHG emissions of corn stover during the growth period are not considered due to its agricultural waste property and to prevent double counting from corn. Full evaporation of wastewater is used to provide process water and steam supplies, considering the weak infrastructure for wastewater treatment and the freshwater supply in agricultural regions. The rigorous calculations show that producing one metric ton of purified L-lactic acid consumes 2.87 ton of corn stover as carbohydrate feedstock, 219.3 kWh of electricity, and 2.98 ton of fresh water for process use with the generation of 1.24 ton of wastewater. No external heat energy input is needed, because the lignin residue combustion provides sufficient heat energy for process use. Based on the detailed data, the calculated GHG emissions for producing one kg L-lactic acid by dry biorefining of corn stover is 0.618 kg CO_2 equiv which is only 18% of that produced by dry milling of corn. This study provides an important sustainability basis and decision-making support for biobased plastic implementation.



	Dry milling of corn	vs.	Dry biorefining of corn stover
L-lactic acid titer (%, w/w)	14.90		12.30
L-lactic acid yield (ton/ton feedstock)	0.480		0.349
Fresh water input (ton/ton product)	2.57		2.98
Wastewater generation (ton/ton product)	1.15		1.24
Electricity consumption (kWh/ton product)	49.46		219.29
Steam consumption (GJ/ton product, total)	21.82 (fossil fuel combustion)		0 (lignin residue combustion)
GHG emission (kg/kg product)	**3.408**		**0.618**

KEYWORDS: *GHG emission, L-lactic acid, corn stover, dry biorefining, wastewater, lignin combustion*

■ INTRODUCTION

Polylactic acid (PLA) plays a significant role as a promising alternative to petroleum-derived polymers, and in reducing greenhouse gas emissions and minimizing their ecological impact.[1,2] As of 2024, the global production capacity of PLA is approximately 9,16,000 t,[3] with 90% derived from grain starch and 10% from other biomass sources such as sugar cane and sugar beet.[4−7] Currently, there is no commercial production of lactic acid from nongrain biomass such as corn stover, wheat straw, and rice straw. The use of nongrain lignocellulose biomass as carbohydrate feedstocks for production of lactic acid and polylactic acid (PLA) is an inevitable choice due to the limited capacity of starch and sucrose production.[2,8] Therefore, the rigorous assessment of greenhouse gas emission reductions and the minimization of ecological impacts by using nongrain lignocellulosic biomass feedstock in PLA production are crucial. Within the PLA production chain, the chiral lactic acid production step is particularly significant in terms of greenhouse gas emissions. Crop cultivation requires fertilizers, pesticides, and electricity; therefore, the production of corn, wheat, or rice grains is generally not regarded as GHG emission neutral or negative. However, this GHG generation generally is not attributed to crop residues such as corn stover, wheat straw, and rice straw as these are considered as agricultural wastes, as well as to avoid double counting of GHG emission from crops.[9,10] Consequently, high-purity lactic acid monomer and PLA produced from lignocellulosic biomass could result in significantly lower or even carbon-neutral GHG emissions compared to those produced from corn.

GHG emission evaluation of chiral lactic acid production from lignocellulose should be based on advanced biorefinery technology, as per the lessons learnt from cellulosic ethanol demonstration facilities. Conventional biorefining technologies are plagued by several critical limitations, including the

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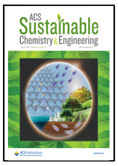

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generation of large volumes of toxic wastewater, excessive energy demands, significant sugar losses, and suboptimal conversion yields in commercial-scale operations.[11] Particularly in the context of producing chiral lactic acid monomers for PLA synthesis, lactic acid derived from lignocellulosic feedstocks frequently falls short of the stringent purity specifications required for effective PLA polymerization.[12−15] In contrast, the dry biorefinery process, which streamlines production by eliminating water-intensive steps, remains underexplored in terms of its greenhouse gas (GHG) emissions profile and broader sustainability implications. To address this knowledge gap, the present study focuses on dry biorefinery technology as the primary processing platform, employing process simulations to quantify the GHG emissions associated with L-lactic acid production from corn stover. This approach is particularly noteworthy as its key performance indicators (KPIs) of conversion yield, water consumption, effluent discharge, and energy consumption are comparable to those of dry milling of corn.[15−19] In the dry biorefinery of corn stover, dry acid pretreatment achieves high-density dry solids with low energy consumption, zero wastewater production, minimal catalyst usage, and low corrosion.[20−22] The biodetoxification process enables rapid and complete biodegradation of high-concentration inhibitors with nearly zero sugar consumption.[23,24] Simultaneous saccharification and cofermentation (SSCF) ensures complete and coordinated utilization of fermentable sugars. This process achieves high-purity (99.6%) L-lactic acid production at a high solid loading by utilizing engineered lactic acid bacteria. The L-lactic acid broth is then purified to polymerization-grade L-lactic acid (optical purity more than 99%) using molecular distillation,[5,25] alongside two other methods with commercial potential: membrane separation[26−28] and solvent extraction.[29−31] Finally, high-purity L-lactide is synthesized using the L-lactic acid obtained from the dry biorefining of corn stover.[15,32]

Rigorous process simulations and greenhouse gas emission evaluation could provide detailed information regarding the technical, ecological, and policy-making supports for the PLA industry. This study starts from a typical lignocellulosic biomass, corn stover, as a feedstock of dry biorefinery processing for production of high-purity L-lactic acid. An Aspen Plus simulation model was established based on dry biorefining design and experimental data with multiple purification options. The energy consumption, freshwater consumption, and wastewater discharge for high-purity L-lactic acid production were analyzed, and the GHG emissions were evaluated by using a life-cycle assessment model under various scenarios. This study provides an important sustainability basis and decision support for industrial implementation for high-purity L-lactic acid production and polylactic acid from lignocellulosic feedstock.

EXPERIMENTAL SECTION

Dry Biorefinery Process for L-lactic Acid Production from Corn Stover or Corn Grain. The process object of this study was the production of purified L-lactic acid by dry biorefinery technology from wheat straw and corn stover.[14,15,17−19,24,33,34] In this study, corn stover was used as lignocellulosic feedstock with a moisture content of 20% and a composition consisting of 35.74% of cellulose, 22.52% of xylan, 17.45% of lignin, and 8.59% of ash.

The annual 3,00,000 t of corn stover processing capacity is the acceptable output for one county, the basic administrative unit in China.[35] The biorefinery plant in this study is modeled as an nth full-scale facility with an annual processing capacity of 300,000 t of dry corn stover or dry corn, equivalent to 37.5 t of dry corn stover or corn per hour with an annual operating time of 8000 h (Supporting Information, Figure S1). The dry biorefinery process of corn stover includes five steps: prehandling, dry acid pretreatment, high solid loading saccharification, submerged liquid biodetoxification, and simultaneous saccharification and cofermentation (SSCF). The corn stover packages are released, the stones, metals, and ash are screened, and then milled to the particle size of ∼1.0 cm in average before pretreatment. The prehandled corn stover is dry acid pretreated with a minimal amount of dilute sulfuric acid solution (with the solid to liquid ratio of 2:1, w/w) and the acid dosage (∼3.5% w/w, based on dry corn stover weight, equal to 35 kg of sulfuric acid per metric ton of dry corn stover). The corn stover and acid solution are cocurrently fed into the pretreatment reactor without any presoaking operation. The saturated water steam (1.6 MPa, 201 °C) is jetted on the corn stover to 175 °C and maintained for 5 min under mild mixing of a helical ribbon stirrer. Then, the pretreated corn stover is quickly discharged without any free aqueous liquid generation.[19,20] The pretreated corn stover is in a dry solid particle form. Then, the solids are enzymatically liquefied into a hydrolysate slurry at a high solid content (30%, w/w) and 50 °C using cellulase enzyme at a dose of 4 mg of total proteins/g of dry corn stover matter (DM).[15,34] After the enzymatic hydrolysis, the hydrolysate slurry is pumped into the bioreactor for submerged liquid detoxification by inoculating *P. variotii* FN89 fungus seed to completely degrade the inhibitors such as furfural, 5-hydroxymethylfurfural (HMF), and acetic acid at a very low sugar consumption (∼2−3%).[23,24] After the biodetoxification, the L-lactic acid fermentation strain *P. acidilactici* ZY271 is inoculated to start the simultaneous saccharification and cofermentation (SSCF) at 45 °C and pH 5.5. The fermentable sugars including glucose, xylose, arabinose, mannose, and galactose in the hydrolysate were fully and coordinately converted into L-lactic acid. After the fermentation, the L-lactic acid broth contains 129.4 g/L of L-lactic acid with high optical purity (99.6%, w/w), which exceeds the requirement of chirality of L-lactic acid monomer for production of chiral lactide.[15]

Dry milled corn is used as control with corn grain at a moisture content of 15% and the composition of 74% of starch, 8.5% of cellulose, 2% of xylan, 9.5% of protein, 4.5% of oil, and 1.5% of ash.[36] Corn grains are milled and liquefied by adding 0.025% (w/w) α-amylase and 0.056% (w/w) glucoamylase at 25% (w/w) of solid loading at 85 °C and pH 5.0,[37] then fermented into L-lactic acid by *Lactobacillus amylophilus* strain.[38]

L-lactic Acid Purification. L-lactic acid purification includes solid−liquid separation, decolorization, acidification, and final refining. The lignin residue solids in the fermentation broth are removed by solid−liquid separation to obtain the clear broth and then decolorized by activated carbon; sulfuric acid is added to form insoluble calcium sulfate and release the free L-lactic acid. Three purification methods are introduced, including the available commercial method of molecular distillation and two methods with commercial potentials of membrane separation and solvent extraction to remove soluble impurities such as residual sugars, proteins, and metal ions to obtain high-purity L-lactic acid. Molecular distillation is carried out at 94 °C and −24.5 Pa vacuum conditions to obtain 92.4% (w/w) of L-lactic acid with the recovery yield of 74.1%[39] (Supporting Information, Figure S2). Membrane separation is performed using microfiltration and nanofiltration, followed by ion exchange and vacuum evaporation to obtain 99.5% of L-lactic acid with the recovery of 76%[26] (Supporting Information, Figure S3). Solvent extraction is carried out using n-butanol and denatured ammonium sulfate to obtain 93% of L-lactic acid with the recovery of 86%[31] (Supporting Information, Figure S4).

The purification step of corn dry milling is carried out by molecular distillation with the same parameters as in the dry biorefinery process.[39]

Full Evaporation of Wastewater (FEW) and Steam Systems. The moisture content of the lignin residue obtained in the solid−liquid separation step was about 55% and it was further dried to a suitable moisture content (∼10%) before being sent to a boiler for combustion. The boiler efficiency for lignin residue combustion was

Table 1. Technical Parameters and Mass Flow of the Dry Biorefinery Process of Corn Stover and the Dry Milling Process of Corn for L-lactic Acid Production

feedstocks	corn (control)	corn stover	corn stover	corn stover
biorefining methods	dry milling[38]	dry biorefining[15]	dry biorefining[15]	dry biorefining[15]
purification methods	molecular distillation[39]	molecular distillation[39]	membrane separation[26]	solvent extraction[31]
experimental L-lactic acid titer in fermentation (g/L)	165.3	129.4	129.4	129.4
calculated L-lactic acid titer in fermentation (g/L)	157.2	131.6	131.6	131.6
experimental purification yield from crude L-lactic acid to purified L-lactic acid (%)	74.1%	74.1%	76.0%	86.0%
calculated purification yield from crude L-lactic acid to purified L-lactic acid (%)	74.0%	73.5%	77.8%	84.5%
calculated purification yield from fermentation broth to purified L-lactic acid (%)	68.1%	66.6%	70.6%	76.6%
experimental L-lactic acid purity (%)	92.4%	92.4%	99.5%	93%
calculated L-lactic acid purity (%)	92.4%	92.4%	99.0%	92.8%
calculated product flow rate (tons/h)	19.423	14.184	14.009	16.222

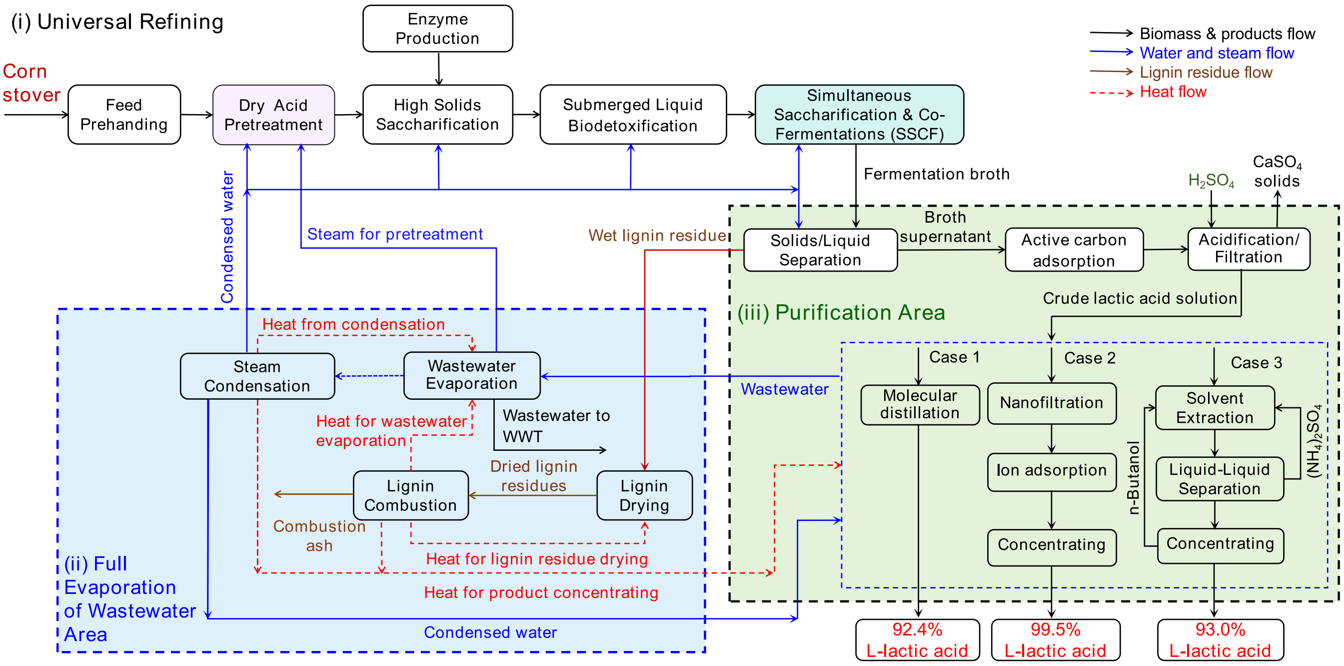

Figure 1. Technical diagram of the dry biorefining process for L-lactic acid production from corn stover. The three areas included are (i) universal refining area including prehandling, dry acid pretreatment, high solid saccharification, biodetoxification, and simultaneous saccharification& cofermentation (SSCF); (ii) purification area including the three options of molecular distillation (Case 1), membrane separation (Case 2), and solvent extraction (Case 3); and (iii) full evaporation of wastewater area by burning the lignin residue to evaporate wastewater and generate hot steam and freshwater for process use.

80%.[33,40] Using lignin residue as a fuel first meets the heat (steam) requirements of the full evaporation of wastewater (FEW) and dry biorefinery processes, compensating for the evaporation process by recovering the heat value through heat-integrated vapor condensation, and then surplus steam is hypothetically directed into a turbine and generator system to produce electricity for use in the plant or for sale to the grid.[41] Steam from the full evaporation of wastewater (FEW) is passed through a condenser to form a clean condensate, which is recycled to the biorefinery steps of pretreatment, saccharification, biodetoxification, fermentation, and purification sections, as an alternative to fresh process water input.[42−44] A small amount of unevaporated concentrated solution is discharged after vapor−liquid separation to conventional wastewater treatment.

Simulation and Technical Settings. The flowsheet simulation model for L-lactic acid production from corn stover or corn grain were developed on Aspen Plus (AspenTech Co, Cambridge, MA) platform with the details shown in Supporting Information. The fundamental data in the flowsheet simulation model was based on the available experimental data from our previous publications, as shown in Table 1. The technical assumptions for the flowsheet simulation model relevant modeling include the following:

(i) The pretreatment step primarily generates inhibitors through the degradation of xylan to furfural, cellulose to HMF, and acetyl groups in hemicellulose to acetic acid. Other inhibitor-forming reactions were not considered.
(ii) Flowsheet simulation of cellulase production was cited from the NREL model for cellulosic ethanol production.[40] The inventory analysis for cellulase was created using the data from Humbird et al.[40]
(iii) Water consumption during equipment maintenance (washing and other operations) was not taken into consideration in this study.

Life-Cycle Analysis and Scenario Studies. This study utilized OpenLCA software (version 2.0.2) and the Ecoinvent 3.9.1 Life Cycle Database to conduct a life-cycle assessment (LCA) of high-purity L-lactic acid (LA) production via a dry biorefinery platform. The

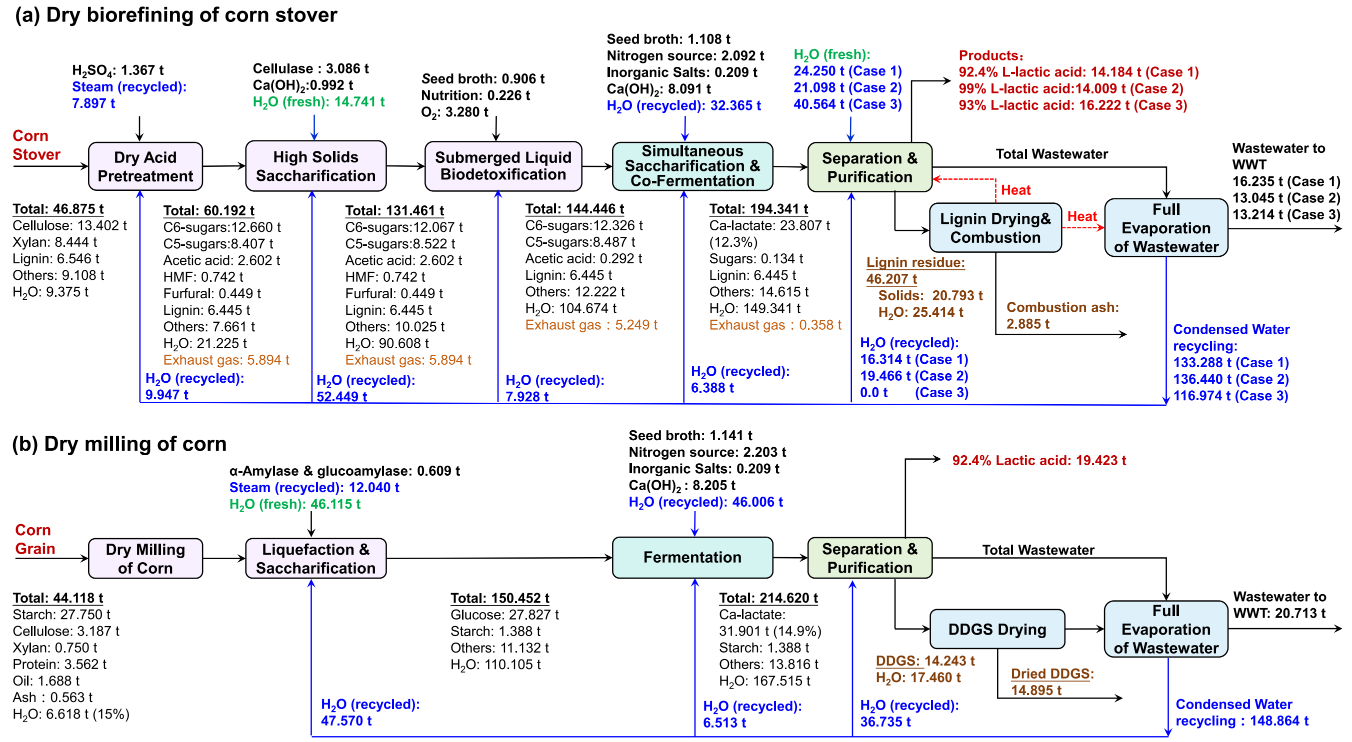

Figure 2. Mass flow rate and water balance of dry biorefining of corn stover (a) and dry milling of corn (b) and for L-lactic acid production. Note: Blue lines, fresh water or condensed water from full evaporation of wastewater; Purification Case 1, molecular distillation; Case 2, membrane separation; Case 3, solvent extraction. The detailed medium composition is shown in Supporting Information Table S2.

ReCiPe Midpoint (H) method was employed to evaluate greenhouse gas (GHG) emissions with 1 kg of L-lactic acid (L-LA) production defined as the functional unit. The system boundary was set at the biorefinery level, starting from the factory gate and ending with the sale of the purified L-lactic acid product at the outlet gate, as shown in Figure 1. The study primarily focused on comparing the environmental impacts of various isolation and purification pathways, specifically quantifying GHG emissions associated with the production of high-purity L-lactic acid. To concentrate on the biorefinery scale,[45,46] upstream processes such as land-use change emissions,[47] soil organic carbon (SOC) changes,[48] and activities related to the harvesting, collection, and transportation of corn stover[49] were excluded from the analysis. GHG emissions during corn growth were attributed to corn grain rather than corn stover, as the latter is categorized as agricultural waste in the Ecoinvent database. This allocation approach avoids double counting, with GHG emissions associated with corn stover growth considered negligible.[50]

This study analyzed greenhouse gas (GHG) emissions under three distinct scenarios for lactic acid production. In Scenario 1 (the base case), all heat (steam) required for the process is supplied by external energy sources, which consist of a mixture of fossil fuels and carbon. Scenario 2 introduces an internal energy source where heat for the dry biorefinery process is generated by combusting lignin residues derived from corn stover as solid fuel. Scenario 3 builds upon Scenario 2 by incorporating additional byproducts: electricity and fertilizers. Specifically, the process recovers phosphorus and potash fertilizers, with one metric ton of dry corn stover corresponding to the recovery of 2.872 kg P_2O_5 and 2.874 kg K_2O fertilizers to the farmland based on the data of 1.6% phosphorus (P) and 3.1% potassium (K) contained in the lignin residue combustion ash (77 kg/ton DM; hereafter, all references to "ton" refer to metric tons).[51] Additionally, the corn dry milling process generates distillers' dried grains with soluble (DDGS) as a byproduct, further contributing to the resource efficiency of the system.

■ RESULTS AND DISCUSSION

Mass Balance of High-Purity L-lactic Acid Production by Dry Biorefining of Corn Stover. The dry biorefinery process starts from the gate of the biorefinery factory and ends with the selling of free L-lactic acid product at the outlet gate, as shown in Figure 1. The mass balance of Aspen Plus modeling for dry biorefining of corn stover and dry milling of corn is shown in Figure 2. In the biorefining section, corn stover undergoes prehandling (size reduction, and dust and solids removal), dry acid pretreatment, high solid loading saccharification, submerged liquid biodetoxification, simultaneous saccharification, and cofermentation (SSCF) to obtain an L-lactic acid fermentation broth with the optical purity of 99.6% (w/w) in the form of calcium salt. No aqueous water phase is generated, and no solid materials are lost in the overall processing chain. In the purification section, the lignin residue solid is removed from fermentation broth by solid−liquid separation; then, the supernatant broth is purified by molecular distillation, membrane separation, or solvent extraction, and the lignin residue is used as solid fuel for combustion to generate heat energy.

The flowsheet simulation modeling for dry biorefining of corn stover is based on an nth-plant factory with the capacity of 3,00,000 t of dry corn stover annually. In the control case, the dry milling of corn is designed with the same feedstock processing capacity of 3,00,000 t of dry corn annually. The Aspen Plus model is built up based on the experimental data as shown in Supporting Information. The experimental and calculated results are shown in Table 1.

The mass balance of Aspen Plus modeling for dry biorefining of corn stover is shown in Figure 2a. In the dry acid pretreatment step, 37.5 tons/h of dry corn stover is mixed with sulfuric acid (1.367 tons/h) and hot steam (7.897 tons/h)

Figure 3. Electricity and steam consumption of dry biorefining of corn stover for production of high-purity L-lactic acid under different purification options. Control, dry milling of corn with molecular distillation purification (a); dry biorefining of corn stover with molecular distillation purification (b), membrane purification (c), and solvent extraction purification (d). The detailed information for the calculations is listed below.

to elevate to 175 °C and maintained for 5 min, of which 85% hemicellulose is degraded to xylose and 20% cellulose is degraded to glucose. In the saccharification step, the pretreated corn stover (60.192 tons/h) is enzymatically hydrolyzed in a viscous liquid hydrolysate slurry (131.461 tons/h, including 11.615 tons/h of glucose, 7.125 tons/h of xylose, 1.397 tons/h of arabinose, 0.179 tons/h of galactose, 0.273 tons/h of mannose, and 1.611 tons/h of unhydrolyzed cellulose) under

Table 2. Detailed Calculations of Electricity and Heat Consumptions

electricity consumptions		
prehandling	pulverization: ∼16.8 kWh/ton; transfer conveyor, conveyor belt, and dust collection system: ∼10.3 kWh/ton	Aden et al.;[52] Humbird et al.[40]
pretreatment	transfer conveyor, pin drum feeder, and plug screw feeder calculated based on Humbird et al.;[36] Mixing system in pretreatment reactor referred to Liu et al.[18] and Hou et al.:[53] ∼7 kWh/ton;	Humbird et al.;[40] Liu et al.;[21] Hou et al.[53]
bioconversion (saccharification, detoxification, and SSCF)	stirring calculated based on the mechanical stirring power formula	Hou et al.;[54] Zhang et al.[55]
equipment operations	pumps, conveyors, pin drum feeders, plug screw feeders, dischargers, and frame-filter compressor based on power consumption data	Humbird et al.[40] (Supporting Information, Table S1)
purification equipment	crystallizer (0.229 kWh/m^3),[56] decolorization (0.375 kWh/m^3),[57] and centrifuge (1.125 kWh/m^3)[58] calculated based on equipment manufacturer's data	Humbird et al.[40] (Supporting Information, Table S1)
dry milling of corn	hammer milling; liquefaction and fermentation	McAloon et al.;[36] Liu et al.[21]

heat consumptions (calculated based on the Aspen Plus model)				
feedstocks	corn (control)	corn stover	corn stover	corn stover
biorefining methods	dry milling	dry biorefining	dry biorefining	dry biorefining
purification methods	molecular distillation	molecular distillation	membrane separation	solvent extraction
pretreatment (GJ/ton of L-lactic acid)	0.00	1.67	1.58	1.46
heater (GJ/ton of L-lactic acid)	3.24	2.15	2.03	1.81
concentrating (GJ/ton of L-lactic acid)	15.85	20.59	23.45	28.14
molecular distillation (GJ/ton of L-lactic acid)	9.83	9.82	0.00	0.00
drying (GJ/ton of L-lactic acid)	1.86	1.22	1.15	1.31
wastewater evaporation (GJ/ton of L-lactic acid)	7.39	7.39	2.61	3.06
total heat consumption (GJ/ton of L-lactic acid)	38.17	42.84	30.83	35.77
heat recovery (GJ/ton of L-lactic acid)	−16.35	−19.79	−15.15	−18.63

high solid loading (30%, w/w). All inhibitory compounds (0.449 tons/h of furfural, 0.742 tons/h of hydroxymethylfurfural (HMF), and 2.602 tons/h of acetic acid) are accumulated in the corn stover slurry since no aqueous water stream or solid stream is generated. In the biodetoxification step, 100% of furfural and HMF, as well as more than 80% of organic acids, are completely biodegraded by the biodetoxification fungus strain with very low sugar loss (less than 2%) to obtain an inhibitor-free fermentable sugar hydrolysate (144.446 tons/h). In the simultaneous saccharification and cofermentation step (SSCF), the fermentable sugars are completely and coordinately fermented into high-chirality L-lactic acid (99.5%) by engineered lactic acid bacterium strain (194.341 tons/h of broth, including 23.807 tons/h of L-lactic acid in the form of calcium lactate and residual sugar less than 1 g/L). In the purification step, the crude L-lactic acid solution (173.382 tons/h) after solid−liquid separation is pumped to molecular distillation to obtain 14.184 tons/h of purified L-lactic acid with a chemical purity of 92.4% (w/w) and a purification yield of 66.6%. The lignin solids (20.793 dry tons/h) are combusted to fully evaporate wastewater into hot steam and condensed water for process use. If molecular distillation is replaced by the other two purification methods, the yield of membrane separation is elevated to 70.6%, but membrane fouling and complexity (microfiltration, nanofiltration, ion exchange, and concentration) in commercial scale should be verified; the yield of solvent extraction is 76.6%, but again, the efficiency and solvent recovery should be verified in commercial scale.

The mass balance for the control case of dry milling of corn is shown in Figure 2b, wherein corn is liquefied and enzymatically hydrolyzed with a solid loading of 25% (w/w) and a starch conversion rate of 95% to obtain the hydrolysate slurry (150.452 tons/h of slurry, and 27.827 tons/h of glucose).[37] In the fermentation step, the sugar is converted to 31.901 tons/h of L-lactic acid in the form of calcium lactate.[38] The fermentation broth (214.620 tons/h) is separated into solids and liquids to obtain crude L-lactic acid broth (199.908 tons/h) and distiller's dried grains with solubles (DDGS) (14.243 dry tons/h) as animal feed. The crude broth is purified to obtain 19.423 tons/h of purified L-lactic acid with a chemical purity of 92.4% by molecular distillation.

The overall water balance shows that the total water consumption for producing 1 ton of L-lactic acid by dry biorefining of corn stover is 13.15 t, of which 2.98 t comes from fresh water and the rest (77.4% of the total) from the condensation of full evaporation of wastewater (Figure 2a). In the control case of dry milling of corn, the total water consumption is 10.87 t per ton of L-lactic acid, of which 2.57 t comes from freshwater consumption and the rest comes from the full condensation of full evaporation of wastewater. Full evaporation of wastewater is used to provide water supply and hot steam to biorefining operations, considering the weak infrastructure for wastewater treatment and freshwater supply in nearby biomass-producing agricultural regions. The heat energy for full evaporation of wastewater comes from the lignin residue combustion in dry biorefinery of corn stover, while the heat in dry milling of corn only comes from the external fossil fuels combustion because the only byproduct from dry milling of corn is DDGS as animal feed. After the full evaporation of wastewater, the final wastewater discharge from dry biorefinery of corn stover is significantly reduced to 1.24 t per ton of L-lactic acid, close to that from dry milling of corn (1.15 t per ton of L-lactic acid).

The total amount of fermentable sugars of dry biorefining (20.590 tons/h, including 56.4% of glucose, 34.6% of xylose, and 9% of arabinose, galactose, and mannose) is smaller than that of dry milling of corn (27.827 tons/h) due to the lower sugar contents in corn stover. This study cited a case of an engineered strain *Pediococcus acidilactici* ZY271 with complete and coordinated conversion of all of the monosaccharides to L-lactic acid (0.636 t of L-lactic acid per ton sugar, or 0.349 t of L-lactic acid per ton of dry corn stover) with the close conversion yield in dry milling of corn (0.645 t of L-lactic acid per ton of glucose, or 0.478 t of L-lactic acid per ton of dry corn grain). Overall, the dry biorefinery of corn stover achieves key performance indicators (KPIs) for conversion comparable to those of dry milling of corn while maintaining low water consumption and minimal wastewater discharge.

Energy Consumption of High-Purity L-lactic Acid Production by Dry Biorefining of Corn Stover. Electricity and steam consumption for L-lactic acid production from the dry milling of corn and the dry biorefining of corn stover are calculated, as shown in Figure 3 and Table 2, based on mass and heat balance, as well as the Mechanical Equipment List in the NREL technical reports.[40,52] Molecular distillation is used as an L-lactic acid purification method for both dry milling of corn and dry biorefining of corn stover. For dry milling of corn, producing one metric ton of high-purity L-lactic acid consumes 49.5 kWh of electricity and 21.8 GJ of steam (Figure 3a), of which the milling step of corn grain accounts for 25% and the molecular distillation step accounts for 43% of the total electricity consumption. The heat for generating hot steam comes from fossil fuel combustion, of which 80% is used for molecular distillation.

For dry biorefining of corn stover, producing 1 metric ton of purified L-lactic acid consumes 219.3 kWh of electricity and 23.05 GJ of steam (Figure 3b), of which the prehandling and pretreatment steps account for 35.4 and 43.6% of the total electricity consumption, respectively. The electricity consumption of dry biorefining of corn stover in the bioconversion steps (saccharification and fermentation) is close to that of dry milling of corn, but the overall electricity consumption for dry biorefining of corn stover is more than four times greater than that of dry milling of corn, primarily due to the high electricity demands of prehandling and pretreatment processes. The overall steam consumption for dry biorefining of corn stover is comparable to that of corn by dry milling, but the heat energy for dry biorefining of corn stover is exclusively generated from the combustion of lignin solids. When membrane separation or solvent extraction techniques are commercially verified, the total electricity consumption could be further reduced by 3 and 12%, and the steam consumption reduced by 23.9 and 25.6%, respectively, compared to molecular distillation (Figure 3c,d).

The heat balance for producing 1 ton of high-purity L-lactic acid by dry milling of corn and dry biorefining of corn stover is shown in Figure 4. Since the primary byproduct DDGS of corn dry milling provides no energy surplus, the heat consumption for molecular distillation (38.167 GJ per ton of L-lactic acid) necessitates an additional 21.815 GJ of heat from external fossil fuels, in addition to heat recovery of 16.351 GJ. In contrast, the total heat consumption of 42.841 GJ per ton of L-lactic acid in molecular distillation of dry biorefining of corn stover is supplied by heat recovery (19.796 GJ) and lignin residue combustion (23.125 GJ), yielding a heat surplus of 0.079 GJ after meeting all heating requirements. If less energy-intensive membrane separation or solvent extraction is used, the total heat consumption is reduced by 28.0 and 16.5%, respectively, compared to molecular distillation. The heat surplus of 6.169 and 2.985 GJ, respectively, is converted into 1162 and 562 kWh of electricity for sale to the grid. The results demonstrate that the efficiency of dry biorefinery of corn stover is significantly superior to that of corn dry milling in terms of heat (steam) consumption.

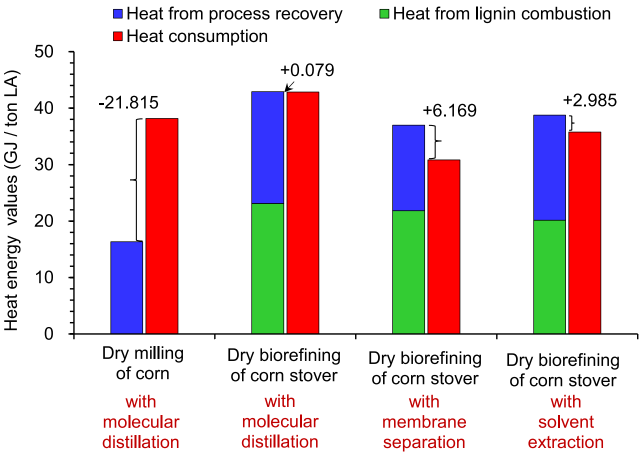

Figure 4. Energy balance of dry milling of corn and dry biorefining of corn stover for production of 1 metric ton of purified L-lactic acid under different purification options. The heat value of lignin residue from corn stover is determined by Liu et al.[33] (~18.878 GJ/ton of dry lignin residue). The heat consumption and heat recovery for overall processing are calculated based on ASPEN as detailed in Table 2.

GHG Emission Analysis for High-Purity L-lactic Acid Production by Dry Biorefining of Corn Stover. GHG emissions by dry milling of corn and dry biorefinery of corn stover for producing 1 kg of high-purity L-lactic acid are evaluated using life-cycle analysis under different scenarios (Figure 5).

For the control case of dry milling of corn, GHG emissions are 4.055 kg of CO_2 equivalent per kg of purified L-lactic acid produced, of which corn cultivation accounts for 37.8% and heat consumption (steam) contributes 55.8% to the total GHG emissions (Figure 5a, Scenario 1). Utilization of byproduct DDGS reduces GHG emissions to 3.408 kg CO_2 equiv for production of 1 kg of high-purity L-lactic acid (Figure 5a, Scenario 3).

For dry biorefining of corn stover, producing 1 kg of purified L-lactic acid generates significantly lower CO_2, 3.039 kg CO_2 equiv (Figure 5b, Scenario 1), mainly from steam heat consumption (78.7% of the total), electricity consumption (7.6%), and cellulase production (6.5%). Utilizing lignin residues as a solid fuel for steam generation (Figure 5b, Scenario 2) results in the sharp reduction of CO_2 emissions to 0.648 kg CO_2 equivalent per kg of high-purity L-lactic acid, indicating that energy consumption control (heat and electricity) is the crucial factor on GHG emissions. Utilizing heat surplus (0.079 GJ/ton of L-lactic acid) for electricity generation and returning phosphorus/potassium from lignin combustion ash to the field (Figure 5b, Scenario 3) further reduce the CO_2 emission to 0.618 kg CO_2 equiv, accounting for only 18% of the GHG emission from dry milling of corn.

The significant CO_2 reduction by dry biorefining of corn stover can be attributed to two main factors: (i) zero CO_2 generation for corn stover due to its classification as agricultural wastes and prevention of double CO_2 counting. The CO_2 generation associated with seed culture, fertilizers, pesticides (herbicides and insecticides), electricity, and fuels is reallocated to corn grain.[9,10] (ii) Heat energy is generated by lignin residue combustion; thus, fossil fuel use is avoided. If less energy-intensive membrane separation or solvent extrac-

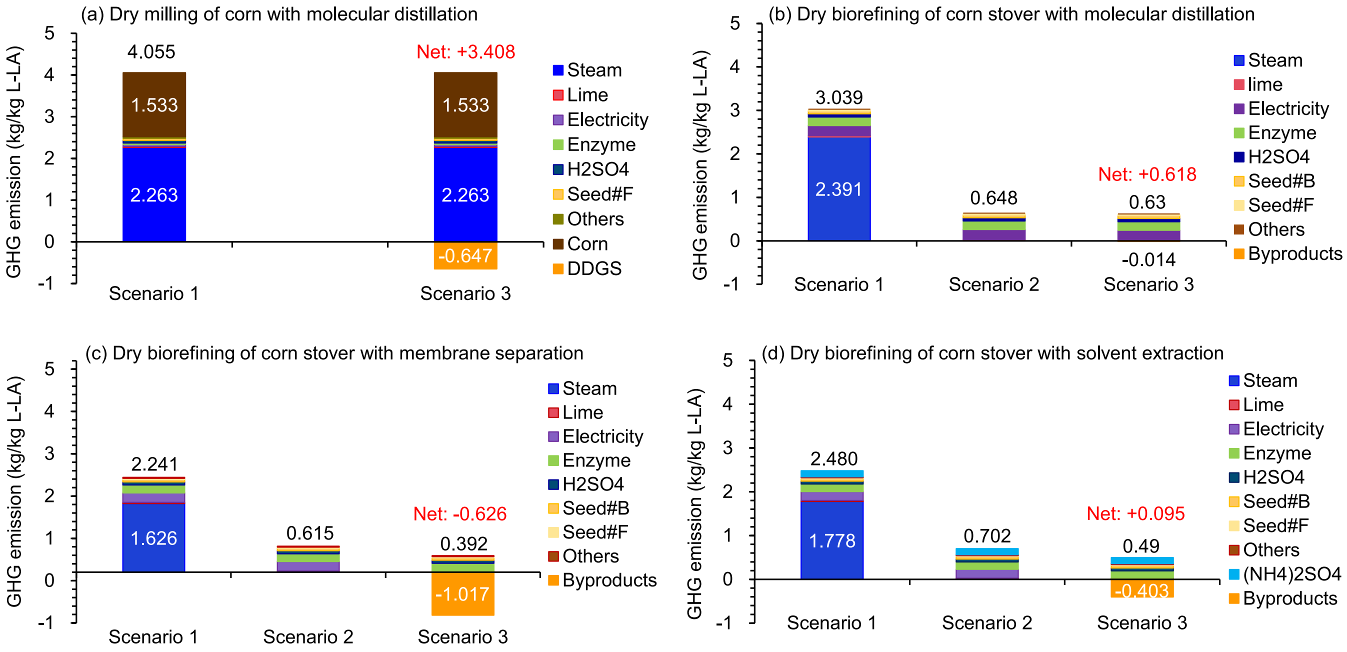

Figure 5. GHG emissions from producing 1 kg of purified L-lactic acid using corn and corn stover; (a) dry milling of corn with molecular distillation; (b) dry biorefining of corn stover with molecular distillation, (c) with membrane separation, and (d) with solvent extraction. Scenario 1, the heat (steam) is supplied by external fossil fuels; Scenario 2, the heat is supplied by combustion of lignin residues from corn stover; Scenario 3, considering electricity generation from lignin residue combustion, and phosphorus and potash fertilizers from lignin combustion ash[51] for Cases 2, 3, and 4 and DDGS generation Case 1. Seed#B refers to detoxification seed culture, Seed#F refers to fermentation seed culture, Others refer to wastewater and gypsum treatment.

tion is used in the purification step, further reduction of GHG emission could lead to the net GHG emissions of −0.626 and 0.095 kg CO_2 equiv, respectively (Figure 5c,d, Scenario 3).

■ DISCUSSION

Energy consumption and the yield of L-lactic acid recovery are the key factors influencing GHG emissions in production of high-purity L-lactic acid by dry biorefinery processing of corn stover, particularly because lactic acid purification significantly affects the yield and quality of the lactic acid product, energy consumption, and GHG emissions in downstream polylactic acid synthesis.[5,59] The traditional crystalline precipitation purification method is difficult to directly achieve the purity standard of polymerization-grade lactic acid products.[12,60] Although esterification distillation techniques create methyl[61] and butyl lactate,[62,63] large-scale ester hydrolysis for lactic acid monomers remains under-validated.[64,65] Alternative methods like electrodialysis,[66,67] adsorption,[68,69] ion exchange,[70,71] and nanofiltration[27,72] have limited industrial validation. This study suggests that low-energy membrane separation and solvent extraction methods could offer advantages in reducing GHG emissions if applied to industrial operations.

The carbon emissions obtained are compared with those reported in the previous publications for L-lactic acid monomer production. When corn is used as feedstock, Vink et al.[73] reported that the GHG emissions by dry milling of corn are 2.26 kg CO_2 equiv for producing 1 kg of high-purity L-lactic acid,[61] compared with 3.408 kg of CO_2 in this study. The key difference is in the estimates of GHG emissions from corn planting: this study uses 1.53 kg CO_2 equiv based on the inventory of Life Cycle Database (Ecoinvent 3.9.1), while Vink et al.[73] uses 0.37 kg CO_2 equiv by considering the GHG fixation during corn growth. If the Vink et al.[73] data of 0.37 kg CO_2 equiv is used, the estimated GHG emissions in this study are 2.24 kg CO_2 equiv, closely aligning with Vink et al.'s[73] reported value of 2.26 kg CO_2 equiv. When lignocellulosic biomass is used as feedstock for high-purity L-lactic acid production, Adom et al.[74] reported a net GHG emission of 0.65 kg CO_2 equiv per kg of L-lactic acid by using corn stover feedstock. Although the value is close to this study (0.618 kg CO_2 equiv), a negative GHG emission for corn stover is adopted in Adom et al.,[74] while this study considers the GHG emission of corn stover is only set to zero. Munagala et al.[75] reported that the GHG emissions from sugar cane bagasse to L-lactic acid is 4.62 kg CO_2 equiv per kg of high-purity L-lactic acid. This value is significantly higher than the current calculation (0.618 kg CO_2 equiv), largely due to the different biorefinery technologies employed: the dry biorefinery process used in this study versus the alkaline pretreatment method used in Munagala et al.[75] Pachón-Rubio et al.[76] reported that the GHG emissions using grapevine twigs feedstock is 3.26 kg CO_2 equiv, with the differences mainly come from the low efficient dilute acid pretreatment and conversion yield, as well as fossil fuels derived electricity, fuel, and the sulfuric acid use. Daful et al.[65] reported that the GHG emissions using sugar cane bagasse and leaves feedstock by is 0.41 kg CO_2 equiv by steam explosion. The low GHG emission mainly contributes to the major CO_2 relocation to the target products of sugar, molasses, and electricity, while the lactic acid byproduct only shared a small percentage of GHG emissions.

The accurate GHG emission evaluation of chiral lactic acid production from lignocellulose should be based on advanced biorefinery technology because the regular technologies used in cellulosic ethanol demonstrations in the past decade are at a low efficiency and certainly will lead to high GHG generations. Li et al.[49] reported a lactic acid production case from corn stover based on the dilute acid pretreatment method[36] with esterification distillation as the purification method and mixed natural gas/lignin residue as the combustion fuel. The process performance by Li et al.[49] is an average or optimal value of many cited cases, instead of a clearly fixed practical process. The calculated GHG emission is 2.79 kg CO_2 equiv, which is several folds greater than the value of 0.618 kg CO_2 equiv in this study. The main reasons for the GHG emission difference between Li et al.[49] and this study come from the different biorefinery technologies used: (i) the conventional dilute acid pretreatment is used by Li et al.[49] under a relatively low solid loading, which generates a higher amount of wastewater and, consequently, a higher heat energy is required; (ii) the inhibitor removal method is not defined and an average value from multiple cited cases is used, which generally requires a high input of fresh water and heat energy; (iii) a rough average of lactic acid production (97.5 g/L) from multiple fermentation cases is used, which is approximately 30% lower than the value (130.3 g/L) in this study; (iv) a rarely used and energy-intensive esterification method is selected for production of free lactic acid; (v) the heat energy is generated by using natural gas (75% of the total fuel), instead of lignin residue byproduct. All of these properties of the process by Li et al.[49] lead to the higher GHG generation than the value in this study.

The analysis above demonstrates that the choice of biorefinery technology and purification process significantly affects the environmental footprint of L-lactic acid production from lignocellulosic biomass. Dry biorefinery technology, when operated with low acid catalyst usage, low energy consumption, and minimal wastewater discharge, can achieve emission levels comparable to those of corn dry processing technology. Moreover, efficient energy integration has led to substantial low-carbon benefits, indicating promising potential for the industrial application of this biorefinery technology.

CONCLUSIONS

This study fills the technical gaps of the detailed energy consumption, water and wastewater balance, and GHG emissions in chiral lactic acid monomer production from nongrain feedstock using a dry biorefinery technology with narrowed performance compared to dry milling of corn. The GHG emissions for producing 1 kg of L-lactic acid by dry biorefining of corn stover using molecular distillation purification is 0.618 kg CO_2 equiv, which is only 18% by dry milling of corn (3.408 kg CO_2 equiv). Using less energy-intensive methods such as membrane separation or solvent extraction for purification could further reduce GHG emissions. This study provides essential technical and sustainability support for the carbon-neutral production of biodegradable polymers from lignocellulosic biomass as alternatives to petroleum-based materials.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.4c10527.

> Aspen process simulation of the dry biorefinery (including separation process and equipment), details of electricity and heat consumption calculations (PDF)

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Y.W.: Conceptualization, Modeling, Validation, Data curation, Investigation, Writing – original draft. B.Z.: Supervision, Visualization, Data curation, Writing – review and editing. J.B.: review and editing. All authors contributed to the revisions of the manuscript.

Notes

The authors declare no competing financial interest.

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