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Research Paper

Balanced water and heat energy recycling by full evaporation of wastewater (FEW) in dry biorefining processes of lignocellulose biomass

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ABSTRACT

Lignocellulosic biorefinery technology requires minimum energy consumption and wastewater generation to overcome challenges in industrial applications. This study established a rigorous model and a comprehensive physical property database of dry biorefining process on Aspen Plus platform for production including L-lactic acid, citric acid, sodium sugar acids, amino acid, and ethanol based on the experimental data. Full evaporation of wastewater (FEW) approach was proposed to completely replaced the external steam supply, and significantly reduced the freshwater input by 67% ~ 85% and wastewater generation by 64% ~ 89%, depending on the specific products. The carbon–neutral heat energy from lignin residue combustion generates an extra heat output of 1.098 ~ 4.772 GJ per ton of dry wheat straw (DW) after all the heat energy needs of the biorefinery process and FEW treatments are satisfied, equivalent to a reduction of 0.219 ~ 0.952 kg CO_2 eq/kg DM emission. This study provided a self-consistent solution for water and energy balance in biorefinery processes.

1. Introduction

The successful deployment of biorefinery technology utilizing lignocellulose feedstock should attain key performance indicators (KPIs) equivalent to dry mill technology using corn feedstock with the minimum wastewater generation and energy consumption (Liu & Bao, 2017a; Liu et al., 2018; Zhang et al., 2021). The low bulk density property and geographical dispersion of lignocellulose resources inevitably lead to small plant size and the plant location close to rural areas where the biomass was produced (Han et al., 2021; Liu & Bao, 2019a). Under this layout, the cost of wastewater treatment and energy supply will be sharply increased due to the inadequate infrastructure system in rural areas, especially in developing countries (Li et al., 2023; Marami et al., 2022). On the other hand, biorefinery technology generates a considerable amount of high-heating value lignin residue as a byproduct. This differed from industrial fermentation technology using corn feedstock with DDGS byproduct as animal feed and no heat value byproduct generation. This high-heating value lignin residue could serve as a solid fuel for multi-effect evaporation of wastewater streams into hot vapor steam and clean condensate water, presenting a significant potential for recycled utilization of wastewater back to the biorefinery chain (Alkasrawi et al., 2013; Li et al., 2019; Zheng et al., 2020; Shao et al., 2023). This concept has been experimentally investigated in terms of the condensation and recycling of the vapors generated by wastewater evaporation for biorefinery process use. However, only a draft principle was proposed and preliminary experimental investigation was conducted without considering the heat balance between the lignin residue combustion and the evaporation (or multiple-effect evaporation) requirement, as well as the water balance between the condensed water recycling and the process water consumption using cellulosic ethanol production as the case (Alkasrawi et al., 2013; Fernandes et al., 2017; Ribeiro et al., 2022; Tobin et al., 2020).

The essential conditions for direct wastewater evaporation in biorefinery processes require that the wastewater be of minimal quality and non-volatile compounds to avoid secondary pollution. The dry biorefinery processing data were used with minimal wastewater generation and fermentable sugar loss, as well as the KPIs were comparable with those of corn dry mill technology (He et al., 2014; Liu et al., 2018; Yi et al., 2019; Zhang et al., 2021). The volatile components in fermentable sugars were eliminated in biodetoxification, fermentation, and purification steps of the dry biorefinery chain (He et al., 2022), providing crucial support to the direct evaporation of wastewater operation.

Rigorous design and validation on Aspen Plus platform based on the experimental data will provide solid and detailed support for the direct

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evaporation of wastewater in dry biorefinery process. The Aspen Plus modeling on the rigorous thermodynamic basis for producing typical fermentation products needs to overcome two key obstacles: (i) Lack of a complete physical property database for biorefinery components (Hantoko et al., 2019). The available physical property database in the latest Aspen Plus V11 system does not cover the full components involving lignocellulose biorefinery processing (Wooley & Putsche, 1996). Filling the physical property gaps of the missing components in biorefinery processes was still a crucial task. (ii) Lack of full flowsheet models to cover the general cases of biorefinery for chiral lactic acid (used as PLA monomer), amino acids, citric acid, and sodium sugar acids (Alves De Oliveira et al., 2019), especially lack of strict downstream processing cases (Islam et al., 2020).

This study tested the concept of eliminating wastewater discharge and fresh water input in dry biorefinery processes by utilizing the heat energy generated from the combustion of lignin residues to evaporate the wastewater into clean water and hot steam through the FEW. The established rigorous models of dry biorefining and the FEW on Aspen Plus platform based on the experimental data include the production cases of biofuel (ethanol), PLA monomer (L-lactic acid), commodity chemical (citric acid), fine chemical (sodium sugar acids), and amino acid (glutamic acid). A comprehensive physical property database involving the components of the models was constructed. The water balance of wastewater evaporation condensation and process water consumption, as well as the heat energy balance from lignin residue combustion and the heat requirement for wastewater evaporation were calculated towards a near-zero wastewater discharge and reduces CO_2 mission (Giwa et al., 2022; Li et al., 2021). This study provided a cost-effective and self-consistent solution of both water and internal energy supply for commercial production of various biofuels and biochemical products from lignocellulose biomass under minimal water, heat, and CO_2 emission.

2. Methods

2.1. Lignocellulose feedstock

Wheat straw is used as the lignocellulose feedstock for Aspen Plus modeling, which contains 20% (w/w) of water, as well as 35.74% of cellulose, 22.52% of xylan, 17.45% of lignin, 4.72% of arabinan, 0.62% of galactan, 0.94% of mannan, 3.00% of protein, 0.34% of extractives, 1.47% of glucose, 2.42% of fructose, 2.20% of acetate, and 8.59% of ash on the dry base (Guo et al., 2023). Wheat straw was milled, and cleaned to remove impurities to a particle size range of 0.1–1.0cm.

2.2. Simulation and software

Aspen Plus V11 (AspenTech Co, Cambridge, MA) was used as the simulator with full authorization from ECUST. According to data from the National Bureau of Statistics of China, the grain output in China for 2023 amounted to 695 million tons (source: https://www.stats.gov.cn/). Based on estimates of agricultural residue output in China, it is a reasonable and minimum projection that each county, the basic administrative unit in China, produces approximately 300,000 tonnes (dry basis) of agricultural residues annually. This figure excludes uses such as animal feed, farmland return, and direct combustion (Liu et al., 2015; Liu & Bao, 2019). Therefore, the process was based on a biorefinery biorefinery plant with a feedstock processing capacity of 300,000 metric tons of dry lignocellulosic feedstock annually. The annual operating time was 8,000 h, equivalent to processing 900 tons of dry feedstock per day.

OpenLCA (version 2.0.2) with the Ecoinvent 3.9.1 Database was used for the Life Cycle Analysis (LCA) and CO_2 release evaluation. The “Global warming” impact assessment methodology was used to assess carbon emissions using hard coal, natural gas, and lignin residues for the FEW.

2.3. Biorefinery process design

Dry biorefining technology was used as the simulation process technology on the Aspen Plus platform. The process parameter settings were adjusted based on the experimental results. The overall process was classified into four areas including universal refining area, fermentation area, purification area, and FEW as shown in Fig. 1. The universal refining area converts lignocellulose feedstock to fermentable sugar syrup. The fermentation area converts the lignocellulose into monosaccharide sugars for fermentation or simultaneous saccharification and co-fermentation (SSCF) into different fermentation products. The purification area removes the inert impurities of lignin residues, residual sugars, proteins, microbial cells, organic byproducts, residual phenolics, minerals, and metabolites from the products. The FEW area evaporates the wastewater into saturated vapor streams by multi-effect evaporation and condensates into pure water for recycled process use.

2.4. Bioconversion of wheat straw feedstock to fermentation products

The biorefining conversion area includes the microbial fermentations using the sugars obtained during the enzymatic hydrolysis (liquefaction) and the simultaneous saccharification together with the fermentation process. Five commodity fermentation products used as biofuels and biochemicals were included in the biorefining conversion area based on the experimental results including lactic acid, citric acid, sodium sugar acids, glutamic acid, and ethanol. The key fermentation parameters were derived from the bench or pilot scale dry biorefining tests and design simulation of the purification process suitable for lignocellulosic systems. For lactic acid, citric acid, and glutamic acid (Cases 1, 2, and 3), an intensive solid–liquid (S/L) separation and an intensive decolorization were required to remove solid lignin residue and the lignin derived melanin precursors, respectively. The fermentation broth was heated to denature and coagulate the suspended proteins and microbial cells and reduce the viscosity to facilitate the subsequent filtration (Datta & Henry, 2006; Meng et al., 2020). The clear supernatant liquid was decolorized by activated carbon adsorption for further purification. The lignin residue solids were sent to the drying unit and the boiler to generate heating energy. After S/L separation and decolorization, the fermentation broth was purified separately to remove residual impurities based on the physical properties of the different products (Fig. 2).

Case 1 (**Fig. S1**) is L-lactic acid production case using an engineered lactic acid bacteria *Pediococcus acidilactici* ZY271 by simultaneous saccharification and co-fermentation at 42 ℃, and pH 5.5, cellulase dosage of 4 mg total cellulase proteins per gram of dry wheat straw to obtain a high optical purity (99.6%) and high concentration (129.4 g/L) of L-lactic acid (He et al., 2022). The fermentation broth was subjected to solid–liquid separation and decolorization to obtain a clear liquid, then evaporated and concentrated for the first crystallization tank at 10°C. The mother liquor from the centrifugation was sent to a double-effect evaporator to concentrate in the secondary crystallization to recover the residual calcium lactate. The crude calcium lactate crystals were washed with water and centrifuged; then the washing water was sent to the first crystallization tank for further recovery of calcium lactate. The crude calcium lactate crystals were sent for acidification using a sulfuric acid solution (50%) and then sent for secondary decolorization using 0.5% activated carbon powder (Alves De Oliveira et al., 2019; Min et al., 2011). The free lactic acid solution was subjected to double-effect evaporation and ion-exchange resin to remove water and metal ions (such as Ca^{2+}, Fe^{2+}, Fe^{3+}, Na^{+}) into the L-lactic acid product of 88% (w/w).

Case 2 (**Fig. S2**) is citric acid production from using *Aspergillus niger* SIIM M288 from corn stover at a solid content of 30% with a cellulase dosage of 6 mg total protein per gram of dry wheat straw feedstock (DM) at an inoculating of 10%, 35 ℃ and pH 4.8 to produced 136.3 g/L of citric acid (Hou & Bao, 2018). The fermentation broth was filtered using

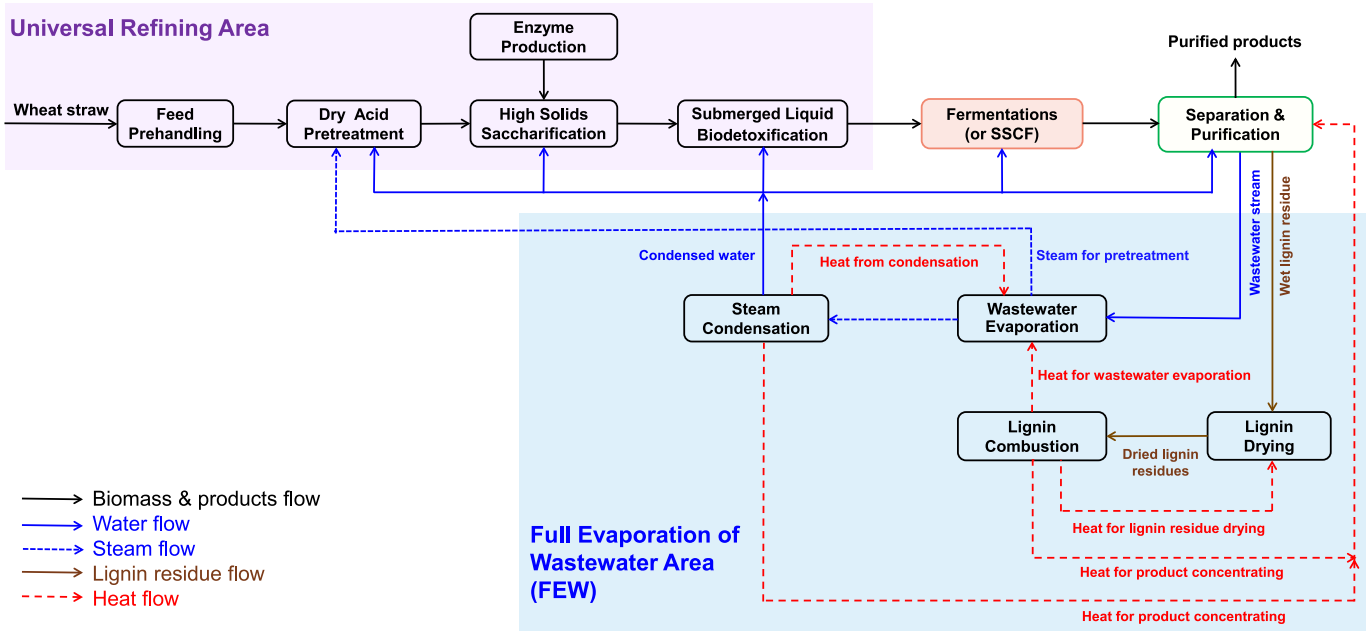

Fig. 1. Illustration of dry biorefinery processing chain and full evaporation of wastewater (FEW) on Aspen Plus platform.

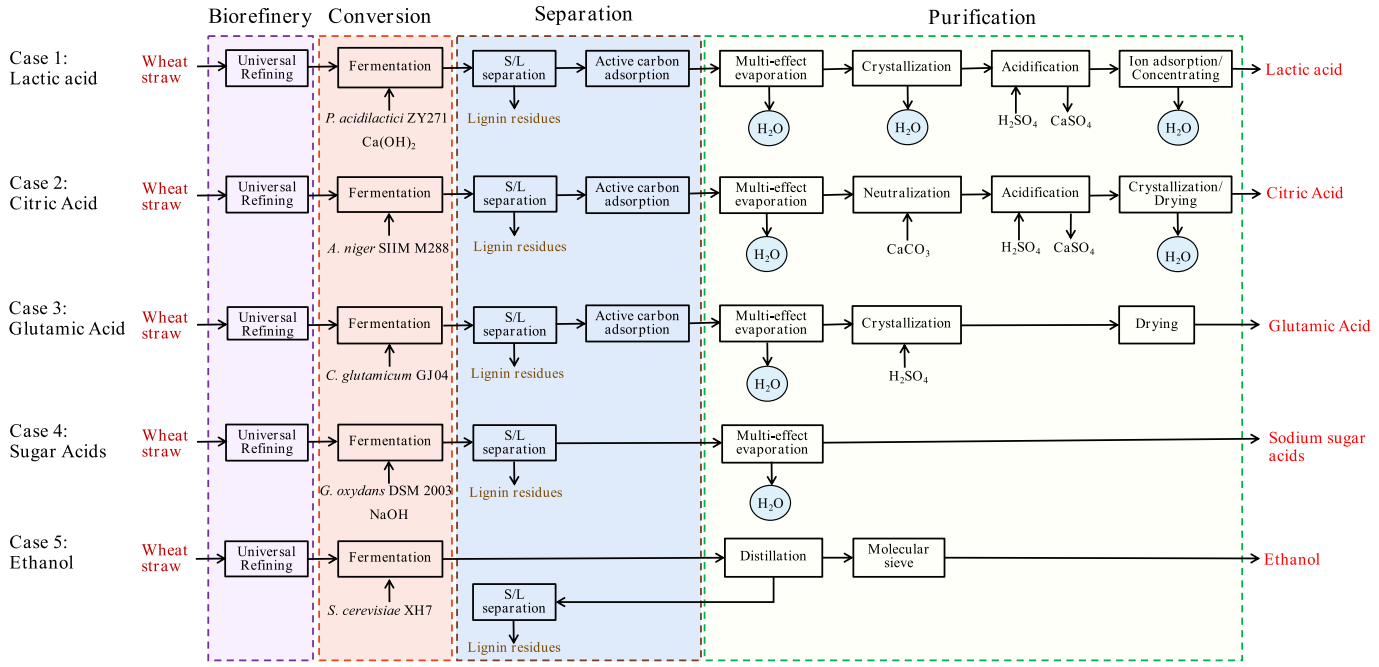

Fig. 2. Process diagram of dry biorefinery chain for production of representative fermentation products from lignocellulose feedstock with a focus on purification steps. Case 1, L-lactic acid; Case 2, citric acid; Case 3, glutamic acid; Case 4, sodium sugar acids; Case 5, ethanol. The detailed process illustrations and descriptions for the purification of each product were illustrated in Fig. S1-S5).

plate and frame pressure filtration and the liquid was decolored with activated carbon to obtain a clear liquid, then sent for neutralization with $CaCO_3$ to obtain calcium citrate precipitate. An equal mass of water was added to wash the precipitate to remove the soluble organic impurities (mainly carbohydrates) (Mores et al., 2021; Wang et al., 2020; Ewing et al., 2022), followed by the acidification by 50% sulfuric acid solution. A small amount of activated carbon was used for the secondary decolorization and then the activated carbon was recovered by solid–liquid separation for recycled use. The crude citric acid solution was then subjected to ion exchange to remove pigments and metal ions such as Ca^{2+}, then evaporated and concentrated by a double-effect evaporator and crystallization, and finally dried to the citric acid product with a purity of up to 98% (Liu et al., 2016; Mores et al., 2021; Wang et al., 2013).

Case 3 (Fig. S3) is glutamic acid production using recombinant strain *Corynebacterium glutamicum* GJ04 by co-fermentation of glucose and xylose to produced 61.7 g/L of glutamic acid at 32 °C and pH 7.2 (Jin et al., 2020). After solid–liquid separation and decolorization, the fermentation broth was concentrated to 20% by the double-effect evaporator and then pH-adjusted with sulfuric acid to its isoelectric point for crystallization to obtain the precipitation. The crystals were washed with water and centrifuged or filtered, finally dried to obtain

98% pure glutamic acid crystals (Kumar et al., 2014; Özüdoğru et al., 2019).

Case 4 (**Fig. S4**) is sugar acid production from corn stover using *Gluconobacter oxydans* DSM 2003 to obtain 118.9 g/L of sodium sugar acids and 59.3 g/L of xylonic acid in the form of sodium sugar gluconate or xylonate at 35 °C, pH 4.8 (Hou et al., 2018). The fermentation broth was concentrated by triple-effect evaporation to 50% (w/w) after the lignin residue was removed by solid–liquid separation as the cement retarder additive (Hou & Bao, 2019; Li et al., 2018).

Case 5 (**Fig. S5**) is for ethanol production from wheat straw using the ethanol fermentation strain *Saccharomyces cerevisiae* XH7 by simultaneous saccharification and co-fermentation to obtain 78.9 g/L (10.0% v/v) of ethanol at 30 °C, pH 5.5, with a cellulase dosage of 4 mg of total protein per gram of dry wheat straw (Liu et al., 2018). The fermentation broth was distilled in two consequent columns to obtain the concentrated ethanol at the azeotropic point (92.5%), and then dehydrated and concentrated to 99.5 % by molecular sieves as fuel ethanol product.

2.5. Design of full wastewater evaporation (FEW)

The conceptual flowsheet illustration of the FEW is shown in Fig. 1, and the Aspen Plus process simulation is shown in **Fig. S6**. The moisture content of the lignin residue obtained in the solid–liquid separation step was about 55 % based on the experimental data (Liu & Bao, 2017b) and simulated by using SEP; the lignin residue cake was dried to the moisture content (~10%) by the waste hot flue before being sent to the boiler for combustion using the HEATER. The boiler efficiency for lignin residue combustion was 80% (Humbird et al., 2011; Liu & Bao, 2017b). The heat from hot lignin combustion flue gas evaporated the wastewater into saturated steam (1.6 MPa, ~200 °C). Partial steam was recycled to the pretreatment section, and the extra steam was condensed into distilled water and recycled to the pretreatment, saccharification, biodetoxification, fermentation, and purification sections. The FEW only generates a small portion of inorganic salts and residual sugar containing wastewater (concentrated solution) from the bottom of the vapor–liquid separation. The Concentrated solution was defined as the wastewater containing 30 % impurities (mainly inorganic salts and residual sugar) to be discharged into the regular wastewater treatment system of the biorefinery plant.

2.6. Technical settings

The assumptions of the flowsheet simulation in this study include:

(i) The main reactions on inhibitor generation in the pretreatment step include the degradation of xylan to furfural, cellulose to HMF, and acetyl groups in hemicellulose to acetic acid. Other reactions concerning inhibitor generation were neglected.

(ii) The cellulase production was consistent with the NREL model (Humbird et al., 2011).

(iii) The water input and output for equipment washing and other maintenance were neglected.

(iv) The compositions and pretreatment efficiency of corn stover in the experimental cases are assumed same to that of wheat straw.

3. Results and discussion

3.1. Filling the gaps of physical properties in general biorefinery process simulations

The incomplete physical property database was the primary obstacle to Aspen Plus simulation to general biorefinery processes. The physical property data of biorefinery modeling generally come from three sources: the embedded database in Aspen Plus, the NREL biomass database (Wooley & Putsche, 1996), and the new component setup by the users. The latest Aspen Plus V11 system has substantially updated biomass components' missing physical property database. The compilation released by the National Renewable Energy Laboratory (NREL) still stands as the most comprehensive biomass database to date (Wooley & Putsche, 1996). When a new component was encountered outside the scope of the embedded Aspen Plus database and the NREL databases, or a component with missing parameters in the two major sources, a new physical property setup becomes necessary from experimentations, molecular thermodynamics-based estimations, or the available databases.

The input of the minimum physical property parameters of the new solid or liquid components is listed in **Table S1**. The estimated physical property parameters of non-solid components based on the molecular structure to meet the minimum parameter requirements are shown in **Table S2**. **Table S3-S6** cover the names, classifications, physical property parameters, and corresponding references of the missing components of feedstocks, intermediates, and products in the biorefinery processes for the production of chiral lactic acid, glutamic acid, sodium sugar acids (sodium sugar gluconate or xylonate), citric acid, and ethanol. The re-organized physical properties of the biorefinery components were classified into four categories by considering the molecular structures, component types (solid or liquid), molecular formulas, and other characteristics:

(i) Components with the full physical property data available in the embedded Aspen Plus database (**Table S3**) such as glucose, lactic acid, cellulose, and water, etc.;

(ii) Components with partial physical property data are available in the embedded Aspen Plus database but require additional input for the missing data (**Table S4**). For example, sodium sugar acid lacks critical temperature (TC), the heat capacity of an ideal gas (CPIG), and liquid vapor pressure (PLXANT);

(iii) Components with the missing physical property data in the Aspen Plus database, but available in the NREL database (**Table S5**) such as xylan, protein, and enzyme properties (Wooley & Putsche, 1996);

(iv) Components absent in both Aspen Plus and NREL database, a new setup was required such as calcium lactate (**Table S6**).

The components and their corresponding parameters in the Aspen Plus database can be recalled without re-entry or estimation, which supports the general biorefinery process simulations.

3.2. Flowsheet simulation of dry biorefinery chain for various biofuel and biochemical productions

The dry biorefinery process illustrated in Fig. 2 starts with the universal refining area, followed by the fermentation area, and finally the separation and purification areas. The universal refining area converts lignocellulose feedstock into highly concentrated and inhibitors-free fermentable sugar syrup (hydrolysate) for consequent bioconversion to various bioproducts (Fig. 3).

The first step of the universal refining area was dry acid pretreatment operated at the co-currently feeding of 46.875 tons/h of wheat straw feedstock (37.5 tons/h of dry matter) and the sulfuric acid catalyst of 1.367 tons/h (96% sulfuric acid), followed by the jetting of 5.956 tons/h of saturated steam (1.6 MPa, 201 °C). The reactor was maintained at 175 °C for 3 min before the steam was released and the pretreated solid of 57.953 tons/h was discharged without generating any free water streams. Approximately 85% of the hemicellulose was converted into xylose, arabinose, or their oligosaccharides, and 20% of the cellulose was hydrolyzed into glucose, oligosaccharides or inhibitory. The inhibitory substances generated include 0.194 tons/h of furfural, 0.127 tons/h of HMF, 0.805 tons/h of acetic acid, and all the inhibitors were absorbed into the pretreated solids.

The second step of the universal refining area was cellulase-catalyzed hydrolysis of the pretreated wheat straw under 30% (w/w) of wheat straw solids into highly concentrated glucose and other monosaccharides. The cellulase enzyme (3.086 tons/h of cellulase solution, equivalent 4 mg total cellulase proteins per gram dry wheat straw) was added to yield 11.615 tons/h of glucose, 7.125 tons/h of xylose, 1.397

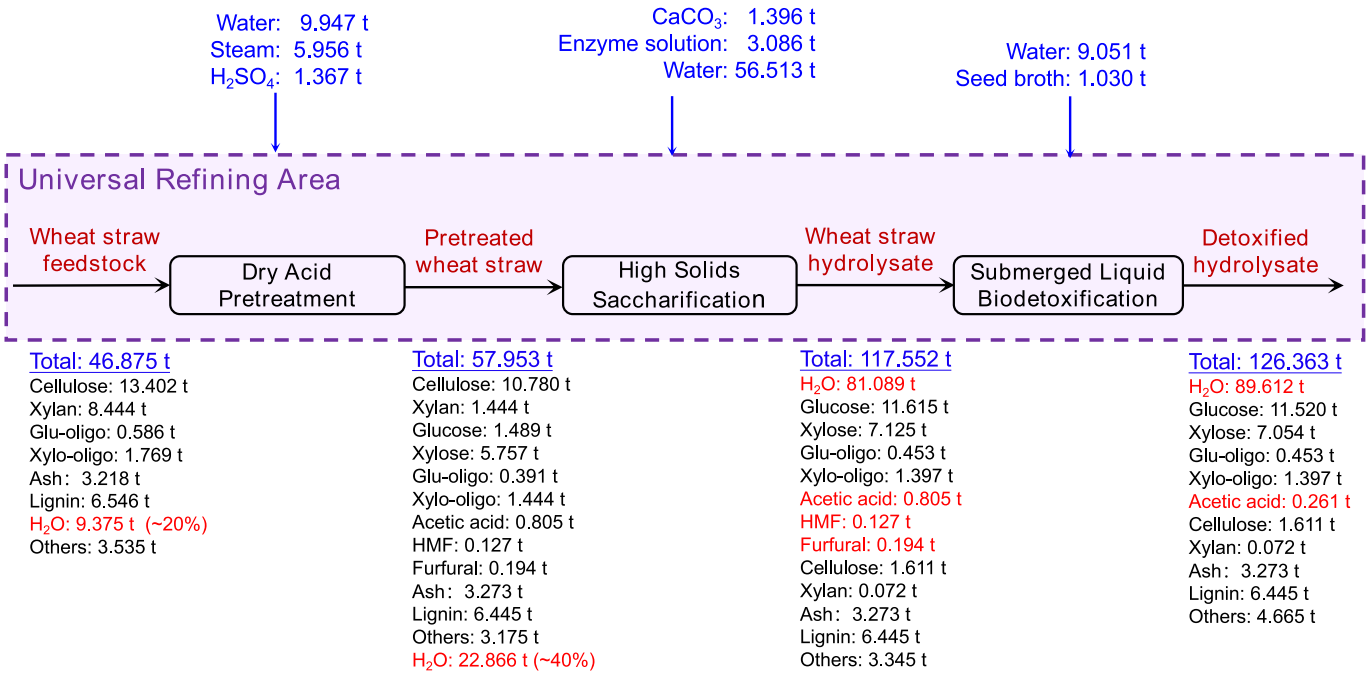

Fig. 3. Mass balance of the universal refining area of dry biorefining of wheat straw.

tons/h of arabinose, 0.179 tons/h of galactose, and 0.274 tons/h of mannose, as well as 6.445 tons/h of insoluble lignin solids and 3.273 tons/h of ash, leaving 1.611 tons/h of unhydrolyzed cellulose for further hydrolysis in the fermentation area (simultaneous saccharification and co-fermentation, SSCF).

The third step of the universal refining area was biodetoxification, providing complete and rapid removal of inhibitors without loss of sugars and extra-streams by inoculating the biodetoxification strain *Paecilomyces variotii* FN89. The furfural and HMF were completely degraded, and 0.261 tons/h of acetic acid (~32% of the total acetic acid) was left in the wheat straw hydrolysate, resulting in the total wheat straw hydrolysate of 126.363 tons/h containing 9% glucose, 6% xylose, 71% water, and 12% insoluble for the consequent SSCF area.

The fermentation and purification areas were summarized in Table 1 based the experimental data. **Case 1** produced 176.245 tons/h of L-lactic acid broth in the form of calcium lactate. Totally 18.681 tons/h of L-lactic acid (88%, w/w), 18.336 tons/h of dry lignin residue solids, and 3.316 tons/h of wastewater were generated after S/L separation, decolorization, crystallization, acidification, ion adsorption, and lactic acid concentrating. **Case 2** produced 139.845 tons/h of citric acid fermentation broth, then the decolored broth reacts with $CaCO_3$ to form calcium citrate precipitate to obtain 13.419 tons/h of citric acid product (98%, w/w) and 18.226 tons/h of dry lignin residue solids after washing, filtration, acidification, crystallization, crystal drying with 15.362 tons/h of wastewater generated. **Case 3** produced 134.828 tons/h of glutamic acid broth and 6.545 tons/h of glutamic acid crystals with a purity of 98% (w/w) after purification with incomplete pentose residues and a slightly higher lignocellulosic residues content (19.291 tons/h). **Case 4** produced 160.280 tons/h of sugar acid broth, 42.766 tons/h of sugar acid (50%, w/w) as a cement retarder additive, and 19.579 tons/h of dry lignin residue solids after simple S/L separation and three-effect evaporation without any wastewater discharge. **Case 5** produced 128.667 tons/h of cellulose ethanol broth, and 8.422 tons/h of 99.5% (w/w) fuel ethanol, 8.273 tons/h of wastewater, and 17.914 tons/h of dry lignin residue solids after regular distillation. A full flowsheet simulation model of dry biorefinery processing was developed, starting from lignocellulosic feedstock to the purified biofuel or biochemical products on the Aspen Plus platform. The result shows that one ton of dry wheat straw produced 0.439 ton of L-lactic acid, 0.351 ton of citric acid, 0.171 ton of glutamic acid, 0.574 ton of sodium sugar acids, or 0.224 ton of ethanol, respectively, while water consumption and reducing high-salinity wastewater discharge were strictly minimized.

Table 1
Technical parameters and mass flow of fermentation steps of dry bi-refining processes for production of typical bioproducts.

	Lactic acid	Citric acid	Glutamic acid	Sodium sugar acids	Ethanol
Fermentation strains	*P. acidilactici* ZY271	*A. niger* SIIM M288	*C. glutamicum* GJ04	*G. oxydans* DSM 2003	*S. cerevisiae* XH7
Fermentation temperature (°C)	42	35	32	30	30
Experimental production (g/L)	129.4	136.3	61.7	177.2	87.0
Sources	(He et al., 2022)	(Hou & Bao, 2018)	(Jin et al., 2020)	(Hou et al., 2018)	(Liu et al., 2018)
Calculated production (g/L)	120.1	130.2	69.8	159.7	78.9
Products flowrate (tons/h)	18.681	13.419	6.545	42.766	8.422
Dry lignin residues flowrate (tons/h)	18.336	18.226	19.291	19.579	17.914
Wastewater output flowrate (tons/h)	3.136	15.362	11.027	0	8.273

Notes: The design study was based on the existing and mature applications of dry acid pretreatment with sulfuric acid as catalyst and the initial solid–liquid ratio of 2:1 (w/w); the high solids loading enzymatic liquefaction was at 50 °C and cellulase dosage of 4 mg total proteins/g DM; the fermentation varies with the specific products as listed in the table; the product titer was based on the grams of the specific products in one liter of liquid fermentation broth (excluding the solids) (He et al., 2014; Zhang et al., 2021; Zhang et al., 2011).

3.3. Water recycling by full evaporation of wastewater using lignin combustion heat

Unlike the starch-derived fermentation with DDGS as byproduct, the biorefinery process generates a large amount of high-heating value lignin residue as byproduct. To achieve a near-zero wastewater discharge in dry biorefinery chain, this study rigorously simulated the FEW on Aspen Plus platform utilizing the heat generated by combusting the lignin residue as the sole source of energy supply. The wastewater contained the least volatile organic compounds after biodetoxification, decolorization, and fermentation, making the direct evaporation of wastewater a viable and feasible option. The hot steam and the condensed water generated from the FEW were recycled back to the required sections of dry biorefinery process, resulting in a complete deletion of external steam input and a significant reduction of external freshwater supply.

Fig. 4 illustrates the water balance of FEW in L-lactic acid case of dry biorefinery process. The total water consumption for processing one ton of dry wheat straw was 5.291 tons, of which 76.7% of the total water consumption (3.899 tons of condensate water and 0.159 ton of vapor steam) was from the FEW operations. The recycled water was used for the universal biorefining area (pretreatment, saccharification, detoxification, 2.014 tons), the multi-stage seed cultures (for fermentation seed, 0.198 ton), calcium hydroxide slurry preparation (0.836 ton), and purification (0.851 ton, shown in blue). Another 9.6% of the total water consumption was from the moisture of raw wheat straw (0.250 ton), cellulase enzyme reagent (0.078 ton), and water generation in bioconversion (0.181 ton). Only 13.7% of the total was from the freshwater input for primary fermentation seed culture (0.054 ton), purification (0.413 ton), and sulfuric acid catalyst solution (0.256 ton). Approximately 1.232 tons of water was lost including 0.3135 ton maintained in the solid wastes (the activated carbon after decolorization, the calcium sulfate after acidification), 0.5975 ton as the moisture in the flue gas of lignin residue drying, 0.1713 ton as the moisture of the air or CO_2 output flows in the biodetoxification and fermentation stages, 0.0325 ton is consumed during the hydrolysis reactions, 0.0588 ton was contained in the final L-lactic acid product (88%, w/w), and 0.0585 ton of water in the concentrated wastewater stream. The overall water balance shows that the recycled condensate water covers 84.9% of the total freshwater consumption and reduces 62.2% of wastewater discharge of the dry biorefinery chain for L-lactic acid production from wheat straw without any outside energy supply. The result reveals that the FEW was both technically and economically feasible for reducing wastewater discharge to fit the biorefinery factory layout in rural areas.

The FEW approaches for reduction of wastewater using the internal energy source of lignin residue combustion were further applied to dry biorefinery processes for production of citric acid, sodium sugar acids, glutamic acid and ethanol cases (Fig. 5**a**). For production of L-lactic acid, citric acid, glutamic acid, sodium sugar acids and ethanol from one ton of dry wheat straw, condensate water through the FEW accounted for 73.7%, 56.2%, 62.1%, 60.1% and 59.7% of the total water consumption, while external freshwater input accounted for 13.7%, 29.3%, 19.0%, 24.1% and 20.4% of the total water consumption, respectively. In Cases 1–3, 92%-96% of the external freshwater input was used in the purification stage. In Case 4, the external freshwater input was mainly in the fermentation stage for preparing sodium hydroxide solution as neutralization reagent (30.1%), and in the purification step as washing water (64.2%). The recycling condensate water fully covered the universal refining stage (pretreatment, saccharification, and biodetoxification) freshwater supply in cases 1–4. Case 5 shows the minimum water consumption (2.830 tons), mainly due to not requiring external freshwater input in the distillation purification step of ethanol. Case 1 shows the maximal water consumption (5.290 tons), mainly due to the alkaline solution preparation in fermentation, lignin residue washing and calcium lactate crystals washing, and acid solution preparation.

The full wastewater evaporation (FEW) concentrates the wastewater stream into a minimal wastewater stream containing 30% (w/w) of inorganic salts and residual sugars, then was sent to the regular wastewater treatment (WWT) area for anaerobic and aerobic digestion (Fig. 5**b**). For producing L-lactic acid from one ton of dry wheat straw (Case 1), the double-effect evaporation and FEW result in a minimal concentrated wastewater discharge and a reduction of 64.4% wastewater discharge due to complete non-glucose sugar conversion with the least residual sugars. For citric acid case (Case 2), 2.377 tons of wastewater was generated due to the washing step to calcium citrate precipitates and citric acid crystals, and the FEW reduces the discharge of concentrated solution to only 0.410 ton (82.8% reduction). For productions of glutamic acid (Case 3) and ethanol (Case 5), the Concentrated solution discharges were 0.294 ton and 0.220 ton (67% and 89% reduction), respectively. Case 4 for sugar acid production achieves a "near-zero waste discharge" of the concentrated solution due to the special properties of sodium gluconate as a cement additive (fermentation broth without further purification). The results indicate that the FEW in dry biorefining process greatly reduces the water demand and wastewater discharge, forming a reasonable water balance with near-zero wastewater discharge.

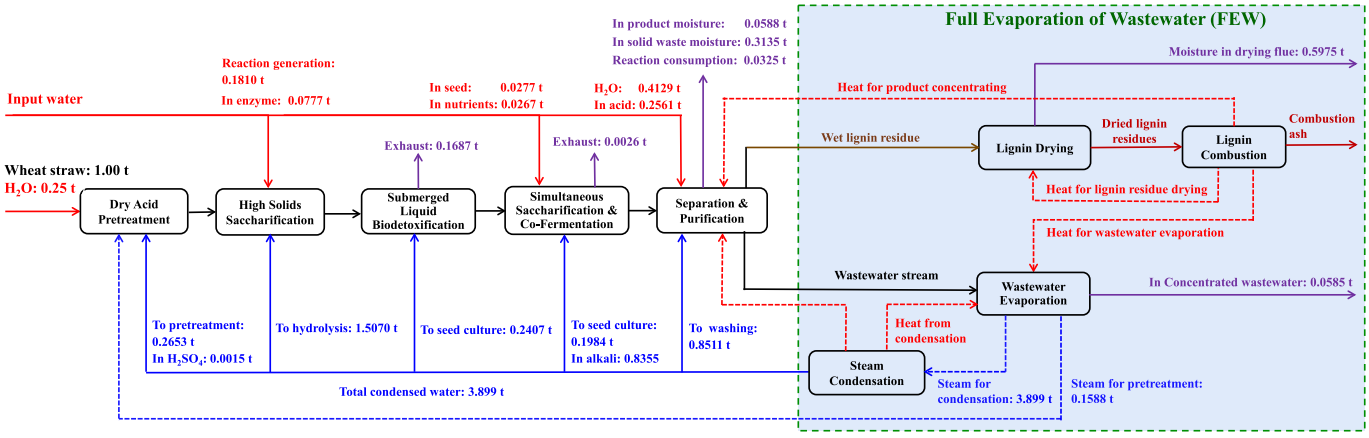

Fig. 4. Water balance per ton of dry wheat straw by the full evaporation of wastewater (FEW) in dry biorefining chain for L-lactic acid production. Red indicates the freshwater input from the external sources, blue indicates the condensate water streams; the purple indicates the non-recoverable water. The freshwater input (red) equals the water in the non-recoverable water (purple). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

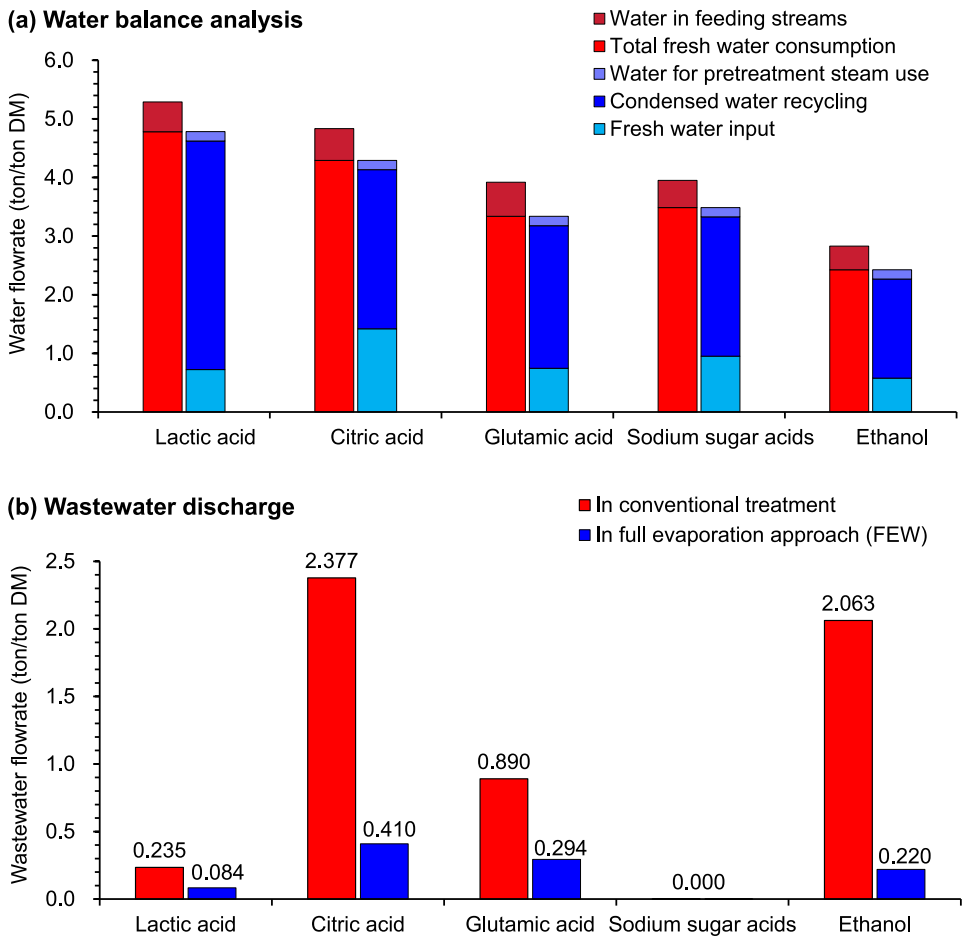

Fig. 5. Water balance analysis and wastewater discharge in dry biorefinery processes with FEW approaches for producing representative biofuels and biochemical products. (a) Water balance analysis, the total water consumption includes total fresh water consumption and water in feeding streams (the moisture of raw wheat straw, cellulase reagents, and water generation in bioconversion; (b) Wastewater discharge; DM, dry wheat straw feedstock.

3.4. Water and heat balance and reduction of greenhouse gas emissions by FEW in biorefinery chain

The heat energy used for FEW comes completely from the combustion of lignin residue generated from the dry biorefining process (Fig. 6).

Fig. 6**a** shows the heat balances of the five biorefinery cases for production of L-lactic acid, citric acid, glutamic acid, sodium sugar acids, and ethanol from one ton of dry wheat straw. Dry lignin residues generated from each case were approximately the same (0.489–0.522 ton) with similar overall heat values (6.961–7.608 GJ). The FEW recovers a maximum of 3.379 GJ (per ton of dry wheat straw) by steam condensation, compensating for the heat consumption of wastewater evaporation. The universal refining area consumes 0.4 GJ for pretreatment, and drying the lignin residue requires 0.37 to 0.39 GJ.

For heat energy consumption in Case 1, the lactic acid concentrating consumes 4.978 GJ and the FEW consumes 0.517 GJ, leaving a 1.098 GJ surplus of heat value. For Cases 2 and 3, the concentrating and crystal drying consumes 2.042 GJ and 2.650 GJ, and FEW consumes 5.207 GJ and 1.579 GJ, leaving 2.428 GJ and 3.204 GJ surplus of heat value, respectively. For Case 4, only the sugar acid concentrating consumes 1.999 GJ, leaving a large 4.772 GJ surplus of heat value. For Case 5, the ethanol distillation consumes 1.454 GJ and FEW consumes 4.584 GJ, leaving 3.360 GJ surplus of heat value. These results show that for the five cases, the heat surplus of 1.098–4.772 GJ from lignin residue combustion can be saved for potential electricity generation after all the process energy requirements are satisfied. Furthermore, the combustion ash of lignin residues contains a considerable amount of phosphorus and potassium elements used to compensate for the loss of phosphorous and potassium to farmland (Liu & Bao, 2019b).

The use of lignin residues from wheat straw biorefining for FEW contributes to the reduction of greenhouse gas emissions (Adom et al., 2016; Pachón et al., 2020). Fig. 6**b** shows the greenhouse gas emissions generated when hard coal (28.9 MJ/kg), natural gas (36.3 MJ/m^3) and lignin residue (18.215 MJ/kg dry lignin residue of wheat straw) were used to provide heat for the FEW process, respectively (Faist Emmenegger M et al., 2007). When hard coal was used as a heat source in FEW, the greenhouse gas emissions were 0.397 ~ 1.124 kg CO_2 eq/kg DM (per kilogram of dry wheat straw) for different bioproducts. The greenhouse gas emissions were reduced by 47.6% using natural gas, which has a higher heat value, compared to hard coal, and the effect of different heat sources on the greenhouse gas emissions was significant. Lignin residue as solid fuel can replace 0.63 tons of hard coal and about 0.36 tons of natural gas (assuming a density of 0.7174 kg/m^3) for heat or electricity generation and no extra CO_2 is generated due to its renewable property of biomass (Sun et al., 2022). The extra heat energy can be utilized to generate electricity and sold to the grid, which was equivalent to the reduction of greenhouse gas emissions of 0.219 ~ 0.952 kg CO_2 eq/kg DM used for electricity production, resulting negative carbon emissions resulting in a significant reduction in the net carbon emissions of the overall dry biorefinery process (Dees et al., 2023).

Advanced biorefinery technologies must be designed to achieve minimal water usage and wastewater generation in order to have the potential for large-scale industrial application (Igbokwe et al., 2022). Commercially hydrothermal, steam explosion, and dilute acid

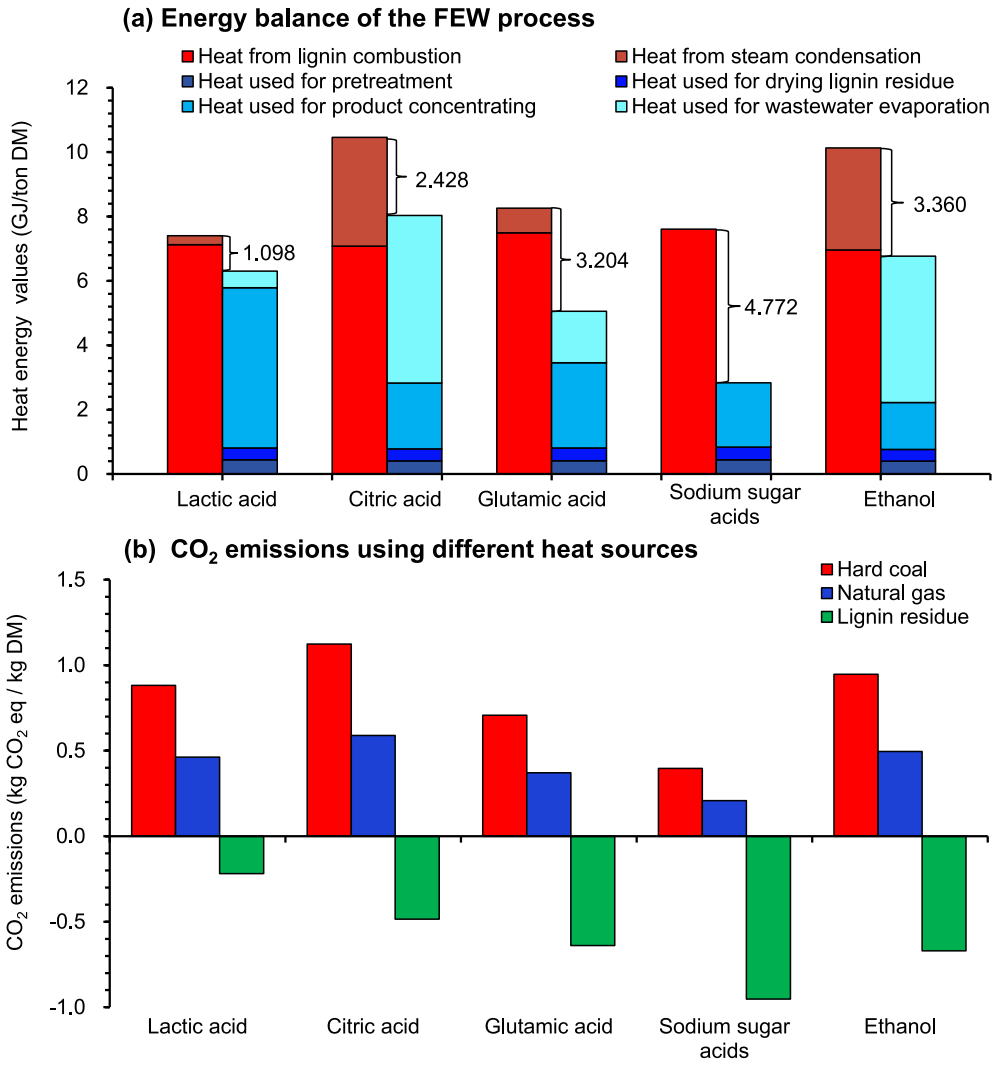

Fig. 6. Energy balance and heat supply greenhouse gas emissions in the dry biorefinery chain with full evaporation of wastewater (FEW). (a) Energy balance of FEW refers to heat consumption of wastewater evaporation, heat demand of pretreatment steam, fermentation broth concentrating (multi-effect evaporation), lignin residue drying; the internal heat refers to the combustion heat of lignin residue and the steam condensation heat (with the efficiency of 80% for both); (b) CO_2 emissions refers to the greenhouse gas emissions in the full evaporation of wastewater (FEW) using hard coal, natural gas, and lignin residue as heat sources, the calculation was based on one kilogram of dry wheat straw matter (DM).

pretreatment methods have been widely used in cellulosic ethanol biorefineries, but the production of each ton of ethanol generates 16 to 115 tons of wastewater containing high inhibitors (Liu et al., 2017; Raj et al., 2022). Wastewater evaporation offers a reasonable and effective solution for wastewater treatment in biorefineries. Palmqvist et al., (1996) were the first to show that evaporated condensate containing volatile components had no negative effect on ethanol fermentation. Alkasrawi et al., (2013) showed that condensate from evaporation in the production of cellulosic ethanol can be used as a substitute for 100% fresh water used in the process. Zheng et al., (2020) tested the evaporation concept by producing cellulosic ethanol through a dry biorefinery platform where the evaporated condensate was recycled to a pretreatment unit. Pretreated wheat straw unchanged in chemical composition, enzymatic hydrolysis, ethanol fermentation performance, ionic content in wastewater, and solid residues, which sufficiently demonstrated that an enclosed circulation of wastewater through evaporation strategy is feasible in dry biorefinery platforms. The FEW processes in this study achieved extremely low freshwater demand and wastewater discharge, and the lignin residue as a solid fuel provided sufficient internal heat demand supply and zero carbon emission advantages. In future work, the dry biorefinery combined with the FEW processes can be extended to a wide range of valuable biobased products including biofuels (ethanol, butanediol, etc.), organic acids (lactic, saccharic, citric, succinic, etc.), and amino acids (glutamic, lysine, etc.), making the dry biorefining process a completely self-consistent and sustainable production technology in terms of water resources and energy consumption.

4. Conclusions

Based on dry biorefinery process with FEW for production of typical biofuels and biochemicals. The incomplete physical property data were supplemented to fill the gap for various biorefinery Aspen Plus simulations. The FEW approaches reduced freshwater input and wastewater generation significantly, using lignin residue provided sufficient heat requirement with a surplus for electricity generation, reducing emissions by 0.219 to 0.952 kg CO_2 eq/kg DM. This study provided a cost-effective and self-consistent solution of both water and internal energy supply for the commercial production of various bioproducts from lignocellulose biomass under minimal water, heat, and CO_2 emission.

CRediT authorship contribution statement

Ya Wang: Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Conceptualization. **Bin Zhang:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Funding acquisition. **Xiucai Liu:** Writing – review & editing, Writing – original draft, Resources, Project administration. **Jie Bao:** Writing – review & editing, Writing – original draft, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2024.12.018.

Data availability

Data will be made available on request.

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