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Case Study

Maximizing cellulosic ethanol potentials by minimizing wastewater generation and energy consumption: Competing with corn ethanol

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GRAPHICAL ABSTRACT

Corn Ethanol *vs.* Cellulosic Ethanol

	Dry Mill	DryPB	DAP	AFEX	DMR	SE
Ethanol titer (%, v/v)	~12%	10.8%	6.8%	5.1%	10.9%	7.7%
Cellulase usage (mg protein/g cellulose)	/	10 mg	20 mg	30 mg	20 mg	25 mg
Wastewater generation (per ton ethanol)	8.332 ton	8.768 ton	16.584 ton	20.712 ton	115.216 ton	115.741 ton
Electricity consumption (per ton ethanol)	34.5 kWh	262.9 kWh	324.6 kWh	529.2 kWh	1028.6 kWh	168.2 kWh
Steam consumption (per ton ethanol)	7.83 GJ	8.63 GJ	11.21 GJ	23.04 GJ	18.60 GJ	11.26 GJ
MESP (per gal ethanol)	~$2.00	$1.79	$2.15	$3.00	$2.56	$3.10

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ABSTRACT

Energy consumption and wastewater generation in cellulosic ethanol production are among the determinant factors on overall cost and technology penetration into fuel ethanol industry. This study analyzed the energy consumption and wastewater generation by the new biorefining process technology, dry acid pretreatment and biodetoxification (DryPB), as well as by the current mainstream technologies. DryPB minimizes the steam consumption to 8.63 GJ and wastewater generation to 7.71 tons in the core steps of biorefining process for production of one metric ton of ethanol, close to 7.83 GJ and 8.33 tons in corn ethanol production, respectively. The relatively higher electricity consumption is compensated by large electricity surplus from lignin residue combustion. The minimum ethanol selling price (MESP) by DryPB is below $2/gal and falls into the range of corn ethanol production cost. The work indicates that the technical and economical gap between cellulosic ethanol and corn ethanol has been almost filled up.

1. Introduction

Sugar platform pathway of lignocellulose biorefining for cellulosic ethanol production includes the steps of prehandling, pretreatment, detoxification (conditioning), hydrolysis, fermentation, and recovery (Lynd et al., 2008). Currently, the biorefining technology is still on the early stage of commercialization with only limited number of commercial scale plants in practical operation (Balan et al., 2013). The concept of minimum ethanol selling price (MESP) is frequently used to quantitatively describe the overall cost of biorefining process starting from the feedstock coming at the factory gate and ending by ethanol product leaving the plant (Aden et al., 2002; Humbird et al., 2011). The MESP values of several mainstream biorefining processing technologies (indicated by their pretreatment methods) are $2.15 per gallon by dilute acid (DAP) (Humbird et al., 2010, 2011), $3.00 per gallon by ammonia fiber explosion (AFEX) (Uppugundla et al., 2014; Kim and Dale, 2015), $2.56 per gal for deacetylation and mechanical refining (DMR) (Chen et al., 2015, 2016), and $3.10 per gal for steam explosion (Chen and Fu, 2016) based on the *n*-th plant assumption with all the required processing technologies at the mature stage. The overall cost of each technology is obviously greater than the current corn ethanol selling price ($2/gal, the average value in the period of 2012–2017, http://www.tradingeconomics.com/commodity/ethanol). Reduction of the overall cost on cellulosic ethanol production to the profitable level

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is strongly required for establishing the full scale industry of bioethanol.

Cost of cellulosic ethanol production mainly comes from three major sections of feedstock, cellulase enzyme, and non-enzyme conversion (Humbird et al., 2011; Liu et al., 2016). Currently, the conversion efficiency indicated by ethanol titer in fermentation broth (5–10%, v/v) (Humbird et al., 2011; Uppugundla et al., 2014; Kim and Dale, 2015; Chen et al., 2016) had been significantly modified and close to the level of corn ethanol production (12–15%, v/v) (McAloon et al., 2000). However, the high conversion efficiency is achieved generally under the prices of high energy consumption and wastewater generation in the step of chemical and inhibitor removal for elevating enzymatic hydrolysis and fermentation yields. The energy and water balance of lignocellulose biorefining is obviously unfavorable to that of the mature dry mill corn ethanol process in consuming higher fresh water, steam, electricity (McAloon et al., 2000; Aden, 2007; Ahmetovic et al., 2010), and generating much more wastewater (Martin and Grossmann, 2011; Sassner et al., 2008; Wingren et al., 2008). Considering the long term goal of cellulosic ethanol as gasoline alternative, the huge fresh water input and wastewater output may create a desperate situation, especially in developing countries such as China, Brazil, and India with the large agriculture industry and biomass production but insufficient fundamental systems. In the wide range of rural area of developing countries, the wastewater treatment system is extremely weak. The pollution to rivers and land by the small local industries has already generated huge environmental problems. It would be a disaster for these regions to implement of a high wastewater releasing industry. Furthermore, electricity supply is still a severe bottleneck for economic activity in many rural regions of developing countries. The huge wastewater generation and energy requirement by the conventional biorefining technology does not fit the reality of the developing countries for cellulosic ethanol industry. Therefore, the reasonable energy consumption and water balance are of special importance for not only it contributes a large portion of the overall cost, but also it determines the feasibility of technology penetration for the coming full scale industry.

This study designed an Aspen Plus model for the dry acid pretreatment and biodetoxification (DryPB) biorefining process. The wastewater generation, the consumption and steam and electricity energy, as well as the MESP value were calculated based on the experimental data and modeling results. The results were compared with several typical biorefining process technologies for cellulosic ethanol production including dilute acid (DAP), ammonia fiber explosion (AFEX), deacetylation, mechanical refining (DMR), steam explosion (SE), and the dry mill process for corn ethanol production. With high ethanol titer, minimum wastewater generation and energy consumption, the MESP of DryPB is below $2 per gal of ethanol, which falls into the cost range of corn ethanol production. The results indicate that the cellulosic ethanol production technology has already significantly advanced to the level for competing with corn ethanol production technology from both technical and economic viewpoint under the proper capital investment, feedstock logistic system and on-site cellulase supply chain.

2. Materials and methods

2.1. Biorefining technologies cited

Five typical lignocellulose biorefining technologies and one corn processing technology for fuel ethanol production were cited in this study. These process technologies are indicated by their unique pretreatment methods including the dry acid pretreatment and biodetoxification (DryPB) (Liu et al., 2017), dilute acid pretreatment (DAP) (Humbird et al., 2010, 2011), ammonia fiber explosion (AFEX) (Uppugundla et al., 2014; Kim and Dale, 2015), deacetylation, mechanical refining (DMR) (Chen et al., 2015, 2016), steam explosion (SE) (Liu and Chen, 2016), as well as the dry mill process for corn ethanol production (McAloon et al., 2000; Wallace et al., 2005). The detailed information is shown in Table 1 and briefly explained as follows:

2.1.1. Dry acid pretreatment and biodetoxification (DryPB)

Corn stover or wheat straw is milled to the size of 10 mm and dry acid pretreated at 175 °C for 5 min using 2.0 g sulfuric acid per 100 g dry biomass (Zhang et al., 2011; He et al., 2014a,b). The dilute sulfuric acid solution and the condensed water are completely adsorbed onto the solids to form 50% (w/w) of the dry pretreated feedstock solids. The pretreated corn stover or wheat straw solids is neutralized to 5.5 by $Ca(OH)_2$ suspension slurry, briefly milled to remove the extra-long fibers, and aerobically biodetoxified at 28 °C and 0.8 vvm of aeration for 36 h (wheat straw) or 48 h (corn stover) (Zhang et al., 2010; He et al., 2016). Xylose and glucose released during the pretreatment are preserved without observable loss. The pretreated and biodetoxified corn stover or wheat straw solids is enzymatically hydrolyzed at 50 °C, pH 4.8 for 12 h at 30% (w/w) solids loading and 10 mg protein/g cellulose of cellulase dosage. The simultaneous saccharification and co-fermentation (SSCF) is performed at 30 °C for 96 h by *Saccharomyces cerevisiae* XH7 (Liu et al., 2017).

2.1.2. Dilute acid pretreatment (DAP)

Corn stover is milled to the size of 4.1–5.8 mm and dilute sulfuric acid pretreated at 158 °C for 5 min using 2.2 g sulfuric acid per 100 g dry biomass. The pretreated corn stover slurry of 30% solid content is flash-cooled and neutralized by ammonia hydroxide. The hydrolysis is performed at 48 °C for 84 h at 20% (w/w) solid loading and 20 mg protein/g cellulose of cellulase dosage. The co-fermentation is conducted by *Zymomonas mobilis* 8b at 32 °C for 36 h.

2.1.3. Ammonia fiber explosion (AFEX)

Corn stover is milled to the size of 0.42 mm and ammonia fiber explosion pretreated at 140 °C for 15 min with 100 g anhydrous ammonia per 100 g dry biomass and 38% (w/w) solid loading. Ammonia and partial water are stripped by steam, then quenched, cooled, compressed, reheated and recycled to the pretreatment reactor (Bals et al., 2011). The ammonia-free biomass is cooled and hydrolyzed at 50 °C for 3 days with the cellulase usage of 30 mg protein/g cellulose and 18% (w/w) solid loading followed by the co-fermentation using *Saccharomyces cerevisiae* 424A at 33 °C for 120 h.

2.1.4. Deacetylation, mechanical refining (DMR)

Corn stover is milled to the size of 19 mm and deacetylated at 80 °C for 2 h using 50 g sodium hydroxide per 100 g dry biomass and 8% (w/w) solid loading. The black liquid is drained overnight. The solid part is again mixed with 12 folds excess of flesh water for 1 h and the residual NaOH is neutralized by adding sulfuric acid. The solids is drained and dewatered to the solid content of 45–50% (w/w). Two rounds of mechanic milling are performed on the deacetylated corn stover and then hydrolyzed at 50 °C for 120 h at 28% (w/w) of solid loading and 20 mg protein/g cellulose of cellulase usage, followed by the co-fermentation using *Zymomonas mobilis* 13-H-9-2 at 33 °C for 22 h.

2.1.5. Steam explosion (SE)

Corn stover is milled to the size of 20–30 mm and steam explosion pretreated at 180 °C for 24 min (Liu and Chen, 2016). The pretreated corn stover of 40% solid content is washed with a 15 folds excess of fresh water and then the solid/liquid separation is performed to obtain the solid (assuming moisture content of the solid is 50% (w/w) due to lack of detail data). The hydrolysis is performed at 50 °C for 12 h with 25 mg protein/g cellulose of cellulase usage and 20% (w/w) solid loading, followed by the co-fermentation using *Saccharomyces cerevisiae* IPE003 at 30 °C for 96 h.

2.1.6. Corn ethanol

Corn grain is hammer milled, continuously liquefied at 88 °C and 20% (w/w) solid loading, then heated to 110 °C for 20 min and saccharified in stirred tank at 60 °C for 6 h. The saccharified corn mash is cooled to 32 °C and fermented at 34 °C and pH 3.5 for 46 h. The ethanol

Table 1
Biorefining performance of the typical biorefining processes for cellulosic ethanol production.

Processing technologies	DryPB		DAP	AFEX	DMR	SE
Pretreatment	Dry acid pretreatment		Dilute acid pretreatment	Ammonia fiber explosion	Deacetylation, mechanical refining	Steam explosion
Final solids content	50%		30%	38%	8%	40%
Temperature and residual time	175 °C, 5 min		158 °C, 5 min	140 °C, 15 min	80 °C, 120 min	180 °C, 24 min
Catalyst and loading (per gram dry biomass)	Sulfuric acid, 20 mg		Sulfuric acid, 22 mg	Ammonia, 1000 mg	Sodium hydroxide, 50 mg	No catalyst
Detoxification	Biodetoxification		Ammonia treatment	No detoxification	Water washing	Water washing
Hydrolysis and fermentation[a]	SSCF		SHCF	SHCF	SHCF	SSCF
Hydrolysis	50 °C for 12 h		48 °C for 84 h	50 °C for 72 h	50 °C for 120 h	50 °C for 12 h
Fermentation	30 °C for 96 h		32 °C for 36 h	33 °C for 120 h	33 °C for 22 h	30 °C for 96 h
Cellulase enzyme and dosage (mg/g cellulose)	CTec2, 10 mg/g		Spezyme CP, 20 mg/g	CTec2 + HTec2, 30 mg/g	CTec3 + HTec3, 20 mg/g	CTec2, 25 mg/g
Feedstock	Corn stover	Wheat straw	Corn stover	Corn stover	Corn stover	Corn stover
Initial solids loading (%, w/w)	30	30	20	18	30	20
Ethanol titer (g/L)	85.1	87.0	54	40	86	61
Ethanol titer (% v/v)	10.8	11.0	6.8	5.1	10.9	7.7
Ethanol yield (%)[b]	84.7	82.8	85.5	63.9	81.0	63.2
MESP ($/gal)[c]	1.79	1.75	2.15	3.00	2.56	3.10
Feedstock	0.87	0.85	0.74	N/A	N/A	N/A
Enzyme	0.19	0.19	0.34	N/A	N/A	N/A
Non-enzyme conversion	0.73	0.71	1.07	N/A	N/A	N/A
References	Liu et al. (2017)		Humbird et al. (2010, 2011)	Uppugundla et al. (2014) and Kim and Dale (2015)	Chen et al. (2015, 2016)	Liu and Chen (2016)

[a] SHCF, separate hydrolysis and co-fermentation; SSCF, simultaneous saccharification and co-fermentation.
[b] The ethanol yield (%) in SSCF was calculated based on the method by Zhang and Bao (2012).
[c] The minimum ethanol selling price (MESP) value of DAP was cited from Humbird et al. (2011); AFEX was cited from Kim and Dale (2015); DMR was cited from Chen et al. (2016). Enzyme cost of DryPB process was calculated based on NREL report which enzyme was produced on site (Humbird et al., 2011); Total cost of ethanol of SE was cited from Chen and Fu (2016).

titer in the fermentation broth is 9% by weight (12%, v/v).

2.2. Aspen plus models

The flowsheet simulation model for DryPB was established on Aspen plus platform (AspenTech Co., Cambridge, MA, USA) based on the NREL model (Humbird et al., 2011) as shown in Fig. 1. The flowsheet includes ten process areas of feedstock handing (A100), pretreatment (A200), detoxification (A250), hydrolysis and fermentation (A300), cellulase enzyme production (A400), product recovery (A500), wastewater treatment (A600), storage (A700), combustor-boiler-turbogenerator (A800), and utilities (A900). The model was varied according to the DryPB process design in pretreatment, biodetoxification, hydrolysis and fermentation, wastewater treatment.

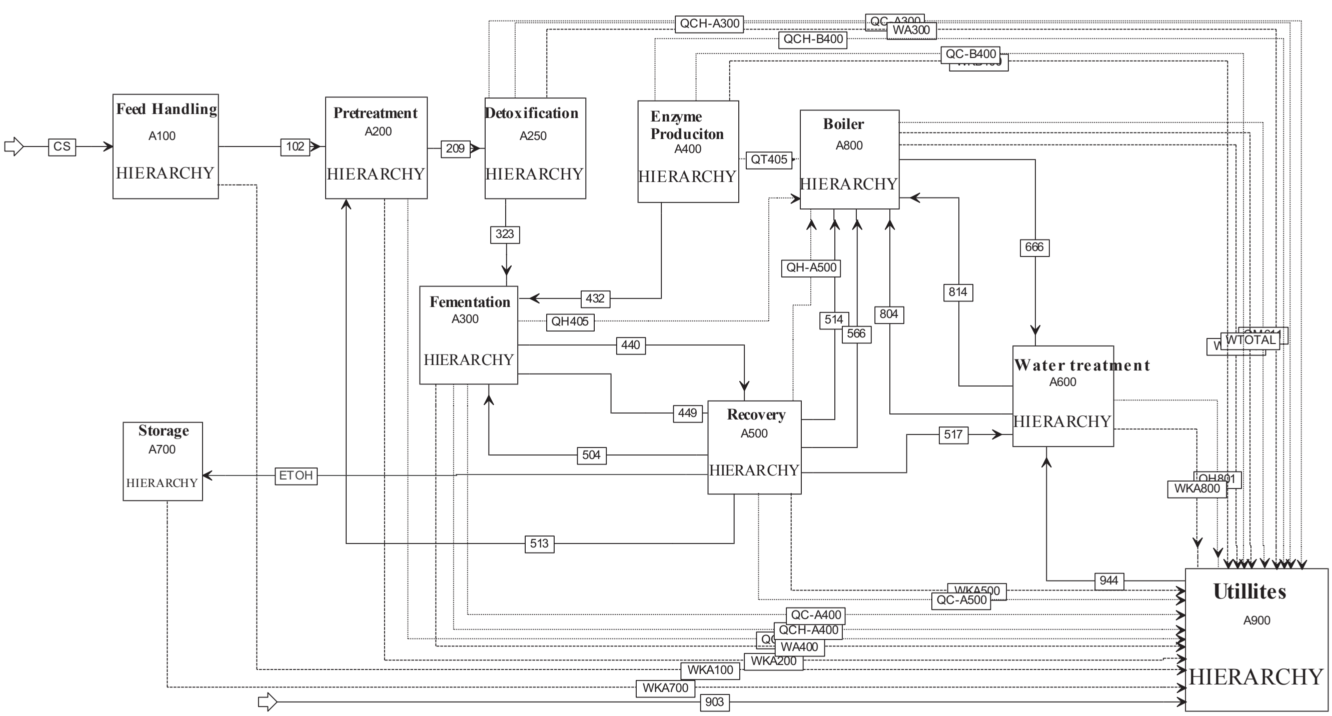


Fig. 1. Diagram of DryPB biorefining process for cellulosic ethanol production.

(1) Pretreatment operation (A200): changed from the conventional dilute acid pretreatment (DAP) in NREL model into the dry acid pretreatment (Zhang et al., 2011; He et al., 2014a,b);
(2) Detoxification (conditioning) operation (A250): separated from the pretreatment area A200 and changed from ammonia overliming in NREL model into the biodetoxification (Zhang et al., 2010; He et al., 2016);
(3) Hydrolysis and fermentation operation (A300): changed from the separate hydrolysis and co-fermentation (SHCF) in NREL model into the simultaneous saccharification and co-fermentation (SSCF). The solids loading of the pretreated and biodetoxified lignocellulosic feedstock is also changed from 20% (w/w) in NREL model to 30% (w/w) solids loading (Liu et al., 2017);
(4) Wastewater treatment (A600): changed by deleting the step of sodium hydroxide (NaOH) addition in the aerobic activated-sludge lagoons for ammonium ions treatment.

2.3. Techno-economic analysis

Biorefining plant size is daily 2000 metric tons of dry lignocellulose feedstock processing capacity and operated for 8410 h annually. The year of 2013 is used as the reference year for adjusting the quotes of equipment, chemicals and labor in a certain year. The exchange rate from US dollar ($) to Chinese Yuan (CNY) is set to 1:6.2 according to the official announcement on the calculation period (http://data.stats.gov.cn/) and the rate may change afterwards. The general equipment of pumps, conveyors and evaporators are quoted from the NREL report (Humbird et al., 2011). The prices of the specific equipment for the DryPB process design in this study including the pretreatment reactor, fermentor and the helical agitator are cited according to the quotes from the related Chinese market.

The total capital investment is calculated based on the total equipment cost, and then variable and fixed operating costs are determined according to plant capacity. With these costs, a discounted cash flow rate of return analysis to determine the minimum ethanol selling price (MESP, $/gal) required to obtain a net present value of zero with 10% internal rate of return after taxes.

The techno-economic analysis data of DAP, AFEX, DMR, and SE are cited from Humbird et al. (2011), Kim and Dale (2015), Chen et al. (2016), and Chen and Fu (2016), respectively. The market sell price of corn ethanol refers to Trading Economics (https://tradingeconomics.com/).

2.4. Wastewater generation and energy consumption analysis

The water and energy balance of the core steps of biorefining processes and corn ethanol production were analyzed based on the published papers or technical reports: DAP process, the NREL technical report (http://www.nrel.gov/extranet/biorefinery/aspen_models/) (Humbird et al., 2011); AFEX, Uppugundla et al. (2014); DMR, Chen et al. (2015, 2016); SE, Liu and Chen (2016); Corn ethanol, the NREL reports (McAloon et al., 2000; Wallace et al., 2005). Corn stover is used as feedstock for all biorefining processes. The core steps of biorefining process included pre-handling, pretreatment, detoxification (conditioning), hydrolysis, fermentation, and recovery. In corn ethanol production process, the alpha-amylase and gluco-amylase are either purchased from outside makers or produced by the on-site enzyme production, similar to cellulosic ethanol production; the wastewater treatment operation is almost the same for both corn ethanol and cellulosic ethanol; the combustion, boiler and power generation operations are for electricity generation and not included in the energy consumption of cellulosic ethanol; the storage and utilities are principally the same for both processes. Therefore, the exclusion of these operations does not give the major impact on the analysis. Besides, if all the downstream processing and recycling of materials and energy are taken into account, the complete water and energy balance in a practical biorefinery plant is too complicated and diverse. Therefore, the on-site enzyme production, wastewater treatment, combustion, boiler and power generation, storage, and utilities are not included for water and energy balance calculation.

The fresh water input refers to the outside water input (excluding the recycled water stream within the process) into the core steps of biorefining processes. The wastewater generation refers to the waste liquid from pretreatment area and the stillage liquid from the beer column of the recovery area after solids/liquid separation.

The energy consumption analysis is referred as the steam and electricity consumption in the core steps of biorefining processes. The areas beyond the core steps such as enzyme production, utilities, storage, wastewater treatment, and boiler, as well as the miscellaneous items such as pump, compressor, conveyer, automation control, washing, heater exchange system are not taken into account for the clarity of energy consumption analysis in the central steps. Steam is consumed on heating the liquid and solids feedstock in the pretreatment and distillation steps, while electricity delivers the power for feedstock size reduction in pre-handling and pretreatment steps, as well as the mixing or driving forces in the steps of pretreatment, hydrolysis, and fermentation.

3. Results and discussion

3.1. Biorefining efficiency of different processing technologies

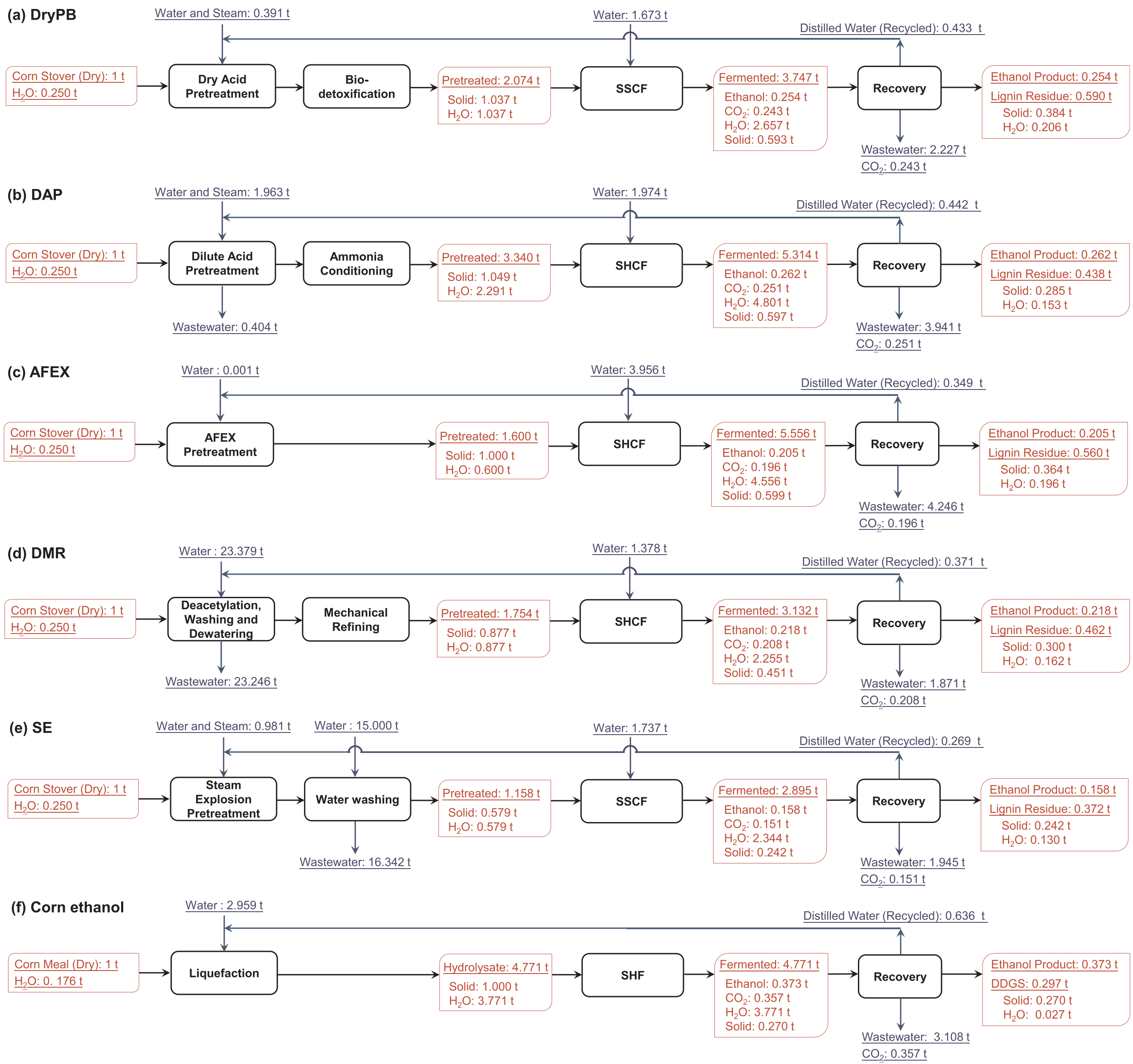
The process parameters and conversion data of DryPB as well as several typical biorefining processes for cellulosic ethanol production are summarized in Table 1. The biorefining process technologies indicated by their pretreatment methods are described in the Methods section. The dry mill technology for corn ethanol production is also included as the target of cellulosic ethanol production (McAloon et al., 2000; Wallace et al., 2005).

In the pretreatment step, DryPB is operated under the highest pretreated biomass solids content (50%), the least pretreatment time (5 min) and chemical dosage (20 mg of sulfuric acid/g DM), comparing to that of DAP (30%, 5 min, and 22 mg/g DM), AFEX (40%, 15 min, and 1000 mg/g DM), and DMR (8%, 2 h, and 50 mg/g DM).

In the detoxification step, the dry acid pretreated solids in DryPB are directly biodetoxified without fresh water and nutrients additions and fermentable sugars loss. As the comparison, DAP uses flash and ammonia conditioning but generate high ammonia ion containing wastewater; AFEX has no detoxification step but the ethanol conversion yield in the consequent hydrolysis and fermentation steps is relatively low; DMR uses water washing and generates massive wastewater; SE also uses water washing and results in the vast wastewater generation and xylose loss.

In the hydrolysis and fermentation steps, DryPB uses the least cellulase dosage (10 mg/g cellulose) to hydrolyze 30% (w/w) of the total solid loading and achieves 85.1 g/L of ethanol (10.8%, v/v) from corn stover, or 87.0 g/L of ethanol (11.0%, v/v) from wheat straw within totally 108 h. As the comparison, DAP uses 20 mg cellulase proteins per gram cellulose to hydrolyze 20% (w/w) of the feedstock and obtains 54 g/L of ethanol; AFEX uses 30 mg/g of cellulase to hydrolyze 18% (w/w) of the feedstock and obtains 40 g/L of ethanol; DMR uses 20 mg/g of cellulase to hydrolyze 30% (w/w) of the total solid loading and obtains 86 g/L of ethanol; SE uses 25 mg/g of cellulase to hydrolyze and ferment 20% (w/w) of the feedstock and obtains 61 g/L of ethanol.

The minimum ethanol selling price (MESP) values for different processing technologies were calculated based on the method by NREL (Humbird et al., 2011). The technical level of DryPB in high ethanol titer (10.8–11.0%, v/v), yield (82.8–84.7%) under the minimum cellulase dosage (10 mg cellulase protein/g cellulose) and high solid loading (50% in pretreatment, 50% in biodetoxification, and 30% in SSCF) leads to the significantly decreased MESP of $1.79/gal (corn stover) or $1.75/gal (wheat straw), comparing to $2.15/gal by DAP



(g) Summary of Water Balance (per ton ethanol)

	DryPB	Corn ethanol	DAP	AFEX	DMR	SE
Water input	8.126 t	7.933 t	15.027 t	19.302 t	113.564 t	112.139 t
Wastewater output	8.768 t	8.332 t	16.584 t	20.712 t	115.216 t	115.741 t

Fig. 2. Freshwater consumption and wastewater generation in the core steps of different biorefining processes. The biorefinery processing technologies are indicated by their unique pretreatment methods including (a) the dry acid pretreatment and biodetoxification (DryPB) process in this study, (b) the dilute acid (DAP) process, (c) the ammonia fiber explosion (AFEX) process, (d) the deacetylation, mechanical refining (DMR) process, (e) steam explosion (SE) process, (f) the dry mill process of corn ethanol (DMP corn), and (g) the summary of overall water balance. The core biorefining areas include pre-handling, pretreatment, detoxification (conditioning), hydrolysis, fermentation, and distillation. The calculation is based on one metric ton of dry corn stover feedstock. One ton of dry corn stover is companied with 0.250 ton of water (20% of moisture). The influence of acid or alkali, nutrient, enzyme, the growth of microorganism and the reactions of water participates on the solid content was not considered in the calculation. The process parameters and results are shown in Table 1. In DMR process, the washing water on the pretreated solids after solids/liquid separation was calculated as the water to solids ratio of 12:1 according to Chen et al. (2015). In SE process, the moisture content of solid fraction after water washing and vacuum filtration is assumed to 50% (w/w) because of lack of detail data. The ethanol yield of DMR and DryPB were based on the ethanol titer (g/L). The ethanol yield of AFEX, DAP and SE were referred to Uppugundla et al. (2014), Humbird et al. (2011) and Liu and Chen (2016), respectively. The generation of waste water in ethanol recovery area was calculated based on the solids loading and final ethanol titer of SSCF or SHCF using the method in the NREL report (Humbird et al., 2011). The recovery area is mainly composed of three columns including the first beer column to concentrate the ethanol to 37% (w/w) as the distillate and remove the dissolved CO_2 in the fermentation broth; the second rectification column is to concentrate the ethanol distillate from the first column to a near azeotropic composition. The rectification column bottoms stream is recycled to the pretreatment reactor as dilution water. The stillage stream of the beer column is filtrated into the wastewater stream and the lignin residue cake containing 35% (w/w) moisture. In the DMP corn ethanol process, corn grain is hammer milled and liquefied then sent for separate hydrolysis and fermentation (SHCF) to yield 12% (v/v) of ethanol. Corn ethanol recovery operation is similar to that of cellulosic ethanol process with the difference in the beer column where the stillage is evaporated, separated and dried to produce the DDGS byproduct (9% of moisture, w/w), instead of lignin residue solids (Wallace et al., 2005).

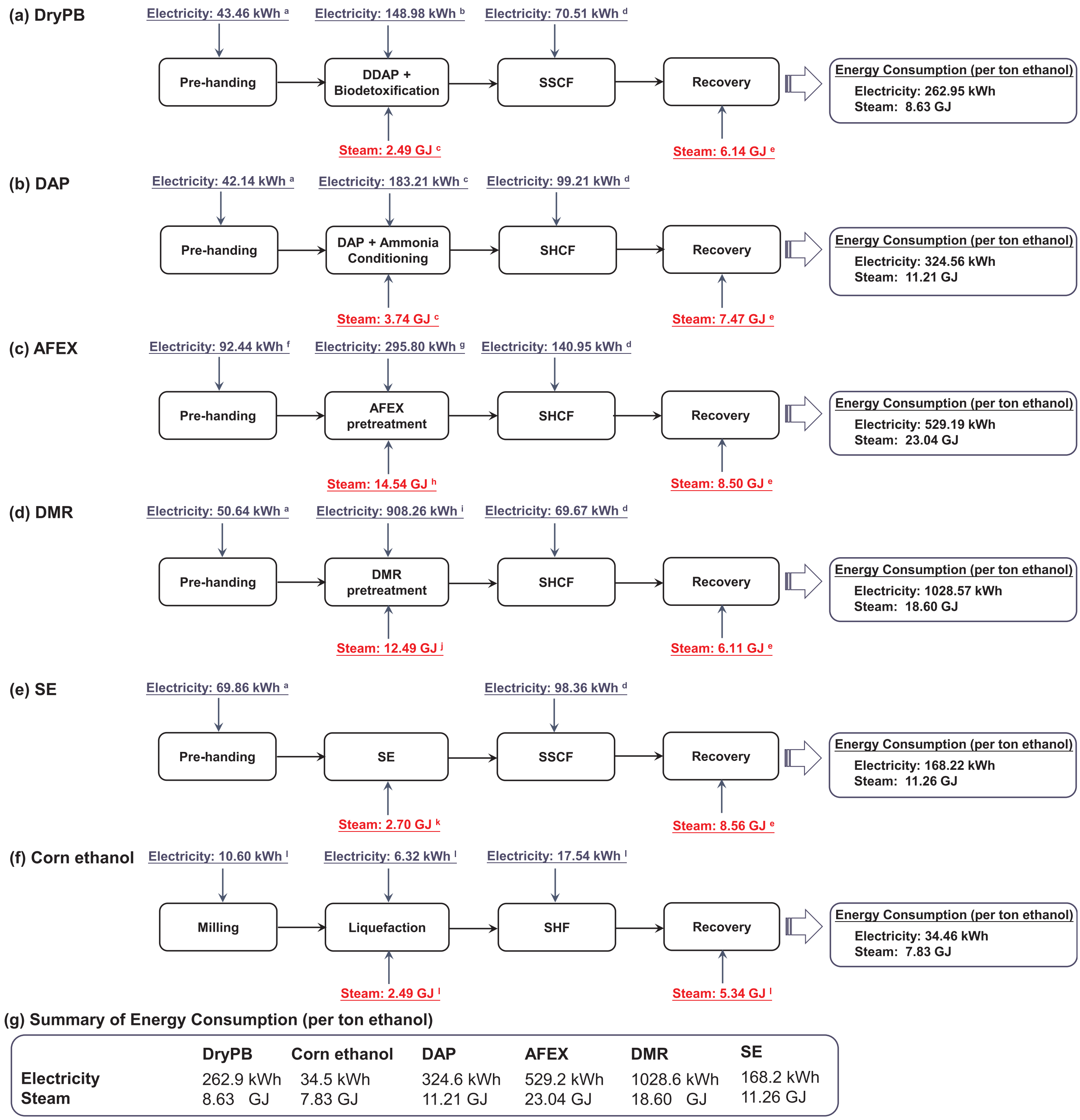


Fig. 3. Electricity and steam consumption in the core steps of different biorefining processes. The core areas include pre-handling, pretreatment, detoxification (conditioning), hydrolysis, fermentation, and distillation. The biorefinery processing technologies are indicated by their unique pretreatment methods including (a) the current dry dilute acid pretreatment and biodetoxification (DryPB) process, (b) the dilute acid (DAP) process, (c) the ammonia fiber explosion (AFEX) process, (d) the deacetylation, mechanical refining (DMR) process, (e) the dry mill process for corn ethanol production, and (f) the summary of the overall energy consumption at the basis of one metric ton of ethanol product. [a]In the pre-handling step of DAP, DMR, and DryPB, the electricity consumption for grinding corn stover into the size of 4.1–5.8 mm in DAP, 19 mm in DMR, 10 mm in DryPB, and 20–30 mm in SE respectively, was calculated based the method by Mani et al. (2004). [b]In the dry acid pretreatment and biodetoxification steps, electricity is consumed on the mixing of the pretreatment reactor and the disk milling of pretreated feedstock. [c]In the DAP pretreatment and conditioning steps, the electricity and steam consumptions are calculated based on the Aspen plus modeling (Humbird et al., 2011). No electricity is consumed on mixing but the electricity for driving the screws is taken into account. [d]In the hydrolysis and fermentation steps of the four processes, the electricity consumption is calculated based on the NREL technical report of 6 W per cubic meter of hydrolysate or fermentation slurry (Humbird et al., 2011). [e]In the ethanol recovery step of the four processes, the steam consumption on beer column and rectification column is calculated by Aspen plus modeling based on the ethanol titer in the fermentation broth. [f]In the pre-handling step of the AFEX process, the electricity consumption for grinding corn stover to 0.42 mm is cited from Lamers et al. (2015). [g]In the AFEX pretreatment step, the electricity consumption comes from the ammonia liquefaction and recycling and the data are cited from Lamers et al. (2015). No mixing electricity is consumed in AFEX pretreatment. [h]In the AFEX pretreatment step, the steam consumption is cited from Stoklosa et al. (2017). The steam consumption for ammonia stripping from the AFEX pretreated corn stover is 2 GJ per metric ton of dry lignocellulose biomass or 9.76 GJ per metric ton of ethanol. The heating steam consumption of the AFEX pretreated corn stover (38% solids, w/w) to 140 °C and 1.72 MPa is 4.78 GJ/metric ton of ethanol based on Aspen Plus modeling. [i]In the DMR pretreatment step, the electricity consumption is on the mechanical refining and cited from Chen et al. (2016). [j]In the DMR pretreatment step, the steam consumption on heating corn stover (8% solids, w/w) to 80 °C was 12.49 GJ per metric ton of ethanol based on Aspen plus modeling. [k]In the steam explosion pretreatment step, the steam consumption on heating corn stover (70% solids, w/w) to 180 °C was 12.49 GJ per metric ton of ethanol based on Aspen plus modeling. [l]In the dry mill process of corn ethanol, electricity and steam consumption is cited from Wallace et al. (2005).

(Humbird et al., 2011), \$3.00/gal by AFEX (Kim and Dale, 2015), \$2.56/gal by DMR (Chen et al., 2016), and \$3.10/gal by SE (Chen and Fu, 2016). The MESP below \$2 per gal is already fall into the range of corn ethanol production cost with \$2/gal of the selling price in the period of 2012–2017 on the international fuel market (http://www.tradingeconomics.com/commodity/ethanol). The result may indicate that cellulosic ethanol by DryPB technology behaves the potential to compete with corn ethanol from both technical and economic viewpoints.

3.2. Freshwater input and wastewater generation analysis on cellulosic ethanol production

Freshwater consumption and wastewater generation in DryPB and other mainstream biorefining processes for processing one metric ton of dry corn stover is illustrated in Fig. 2 and the equivalent data for obtaining one metric ton of fuel ethanol is also summarized.

In the pretreatment and detoxification steps, freshwater is consumed on regulating the chemicals (acid, alkaline, or ammonia) concentration and lignocellulose solids content, as well as to wash the pretreated feedstock for inhibitor removal in some specific pretreatment operations. Wastewater is generated from the operations of solid/liquid separation, steam condensation, or washing. For pretreating and detoxifying (conditioning) one metric ton of corn stover, DryPB consumes 0.271 ton of freshwater and 0.120 ton of steam on adjusting sulfuric acid concentration and solids content but no wastewater was generated. As the comparison, DAP consumes 1.963 tons of freshwater and steam and generates 0.404 ton of wastewater; AFEX, 0.001 ton of freshwater and no wastewater generation; DMR, 23.379 of freshwater and 23.246 tons of wastewater for deacetylation at 8% (w/w) of solids content (11.750 tons of freshwater) and water washing of the deacetylated corn stover (11.629 tons of freshwater); SE, 15.981 tons of freshwater and steam and 16.342 tons of wastewater.

In the hydrolysis and fermentation step, freshwater is consumed on regulating the solids content of hydrolysate slurry and fermentation broth. For processing one metric ton of corn stover, DryPB requires 1.673 tons of freshwater on adjusting the solids content of the hydrolysate slurry and fermentation broth to 30% (w/w) solids (corn stover) content. As the comparison, DAP requires 1.974 tons of freshwater to adjust the solids content to 20%; AFEX, 3.956 tons of freshwater to 18%; DMR, 1.378 tons of freshwater to 30%; SE, 1.737 tons of freshwater to adjust the solids content to 20%.

In the recovery step, the fermentation broth is distillated to 37% (w/w) of ethanol in the beer column and fed to the rectification column to obtain the near azeotropic ethanol composition 92.5% (v/v), then fed to the molecular sieving column to obtain the final fuel ethanol product (99.5%, v/v). The stillage water from the second rectification column is recycled to the pretreatment reactor for partial supplementation of freshwater. The stillage liquid slurry from the beer column is solid/liquid filtrated into the liquid wastewater stream and the solids lignin residue cake. For processing one metric ton of corn stover, DryPB generates 2.227 tons of wastewater and 0.590 ton of lignin residue solids (65% of dry solids) from the stillage stream of the beer column per metric ton of dry corn stover. As the comparison, DAP generates 3.941 tons of wastewater and 0.438 ton of lignin residue; AFEX, 4.246 tons of wastewater and 0.560 ton of lignin residue; DMR, 1.871 tons of wastewater and 0.462 ton of lignin residue; SE generates 1.945 tons of wastewater and 0.372 ton of lignin residue.

Corn ethanol production by dry mill process is similar to cellulosic ethanol production process in the recovery step with the only difference in beer column, in which the stillage is evaporated, separated and dried to DDGS byproduct, instead of lignin residue solids in cellulosic ethanol process. DryPB consumes 8.126 tons of freshwater and generates 8.768 tons of wastewater, which is clearly comparable to 8.332 tons of wastewater generation in dry mill processing of corn ethanol. As the comparison, DAP consumes 15.027 tons of freshwater and generates 16.584 tons of wastewater; AFEX, 19.302 of freshwater and 20.712 tons of wastewater; DMR, 113.564 tons of freshwater and 115.216 tons of wastewater; SE, 112.139 tons of freshwater and 115.741 tons of wastewater.

3.3. Energy consumption analysis on cellulosic ethanol production

Steam and electricity consumption in the core steps of DryPB and other processes are illustrated in Fig. 3 based on Aspen plus modeling. For production of one metric ton of cellulosic ethanol from corn stover, DryPB consumes 2.49 GJ of steam (268 °C and 1.3 MPa) as the heat and freshwater source in pretreatment, and 6.14 GJ of steam (232 °C and 0.95 MPa) to distillate ethanol to 37% (w/w) in beer column and 92.5% (v/v) in rectification column (excluding the steam used on vent scrubber and molecular sieve column). For production of one metric ton of corn ethanol, the dry mill process consumes similar steam of 2.49 GJ in liquefaction of corn meal and 5.34 GJ in distillation. Clearly, the total steam consumption of cellulosic ethanol production in DryPB (8.63 GJ/ton ethanol) is comparable with that of corn ethanol production in dry mill process (7.83 GJ/ton ethanol). As the comparison, DAP consumes 3.74 GJ in pretreatment for heating the moderate solids content (30%, w/w), and 7.47 GJ in distillating 54 g/L of ethanol in the fermentation broth; AFEX, 4.78 GJ in pretreatment for heating feedstock, water and ammonia to 140 °C, 9.76 GJ in stripping ammonia from the pretreated corn stover, and 8.50 GJ in distillating ethanol from the fermentation broth (40 g/L of ethanol); DMR, 12.49 GJ in pretreatment on heating the low solids content slurry (8%, w/w) to 80 °C, and 6.11 GJ in distillating high ethanol titer broth (86 g/L). SE consumes 2.70 GJ in pretreatment for heating the high solids content (70%, w/w), and 8.56 GJ in distillating 61 g/L of ethanol in the fermentation broth.

For producing one metric ton of ethanol, DryPB consumes 43.46 kW of electricity in prehandling on grinding corn stover into pieces with approximately 10 mm in diameter based on the calculation by Mani et al. (2004); 147.64 kWh on disk milling by a disk milling machine (18.75 kW for milling one metric ton of the pretreated corn stover, wet basis); 1.34 kWh is consumed in pretreatment on driving the helical ribbon impeller agitation based on the rheological modeling and CFD calculation on the specific pretreatment reactor by Hou et al. (2016); 70.51 kWh is consumed in prehydrolysis and SSCF on agitating the high solids loading slurry based on the NREL technical report (6 W of electricity is used per cubic meter of hydrolysate) (Humbird et al., 2011). Totally 262.95 kWh of electricity is consumed in the core steps for producing one metric ton of cellulosic ethanol. For corn ethanol production, the dry mill process consumes 10.60 kWh on hammer milling, 6.32 kWh on liquefaction agitation, 2.05 kWh on saccharification agitation, and 15.49 kWh on recirculation pump in fermentation step for producing one metric ton of ethanol (Wallace et al., 2005). Clearly, cellulosic ethanol is disadvantageous over corn ethanol on electricity consumption: 262.95 kWh vs. 34.46 kWh for producing one metric ton of ethanol in the core steps. As the comparison, DAP consumes 42.14 kWh of electricity on cutting corn stover to 4.1–5.8 mm (Humbird et al., 2011) according to Mani et al. (2004); AFEX, 92.44 kWh on cutting to 0.42 mm (Uppugundla et al., 2014; Lamers et al., 2015); DMR, 50.64 kWh on cutting to 19 mm (Chen et al., 2016); SE, 69.86 kWh on cutting to 20–30 mm (Liu and Chen, 2016). In the pretreatment step, DAP consumes 183.21 kWh on the horizontal screw-feeding pretreatment reactor (Humbird et al., 2011); AFEX, 295.80 kWh on ammonia liquefaction and recycling (Lamers et al., 2015); DMR, 908.26 kWh on the two-step mechanical refining after deacetylation (Chen et al., 2016). In the hydrolysis and fermentation step, 99.21 kWh on DAP, 140.95 kWh on AFEX, 69.67 kWh on DMR are consumed for producing one metric ton of ethanol according to the electricity consumption calculation by NREL (Humbird et al., 2011).

Although the electricity consumption of cellulosic ethanol is obviously greater than corn ethanol (262.95 kWh vs. 34.46 kWh), cellulosic ethanol biorefining is a high electricity positive process with extra

Table 2
Case analysis of water and energy balance in biorefining processes.

Biorefining technologies	Freshwater consumption		Wastewater generation		Electricity consumption		Steam consumption	
	Usage (ton/ton ethanol)	Reduction (%)	Generation (ton/ton ethanol)	Reduction (%)	Consumption (kWh/ton ethanol)	Reduction (%)	Usage (GJ/ton ethanol)	Reduction (%)
(a) Under the assumption of the same bioconversion yield with DryPB								
DryPB (Base case)	8.126	/	8.768	/	262.9	/	8.63	/
DAP	7.764	−48%	10.358	−38%	295.9	−9%	9.88	−12%
AFEX	6.496	−66%	8.768	−58%	383.9	−27%	17.88	−22%
DMR	96.403	−15%	100.287	−13%	882.8	−14%	16.86	−9%
SE	99.339	−11%	106.042	−8%	158.2	−6%	8.84	−21%
(b) Under the assumptions of the biodetoxification application and the same bioconversion yield with DryPB								
DryPB (Base case)	8.126	/	8.768	/	262.9	/	8.63	/
DAP	7.764	−48%	10.358	−38%	295.9	−9%	9.88	−12%
AFEX	6.496	−66%	8.768	−58%	383.9	−27%	17.88	−22%
DMR	54.846	−53%	54.551	−53%	263.0	−74%	16.86	−9%
SE	8.126	−93%	8.768	−92%	254.2	+51%	7.82	−31%

Table 3
Impact of enzyme supply mode on the enzyme cost and MESP.

Enzyme supply	MESP ($/gal)	Enzyme cost ($/gal)
On-site production (Base case)	1.79	0.19
Cellulase supply at purchase price of $5.07/kg protein[a]	1.83	0.20
$6.27/kg protein[b]	1.88	0.25
$10.14/kg protein[c]	2.05	0.41
$23.30/kg protein[d]	2.64	0.94

[a] Assume that the cellulase enzyme protein is purchased under the price estimated by Kazi et al. (2010): $507 per metric ton of the enzyme broth containing 10% proteins, equivalent to $5.07/kg (2007$).

[b] Assume that the cellulase enzyme is purchased from Novozymes with the claimed enzyme cost of $0.50/gal ethanol (http://novozymes.com/en/news/news-archive/Pages/45713.aspx). In the proposed process (Humbird et al., 2011), the ethanol production is 21,672.41 kg/h and the enzyme usage is 579 kg protein/h, which are equivalent to 7256.35 Gal ethanol/h (21,672.41 kg/h is divided by the ethanol density 0.789 then transformed to gal). The enzyme protein price is equivalent to 7256.35 0.50/579 = $6.27/kg (2010$).

[c] Assume that the cellulase enzyme protein is purchased under the price estimated by Klein-Marcuschamer et al. (2012) on the fungal cellulase production: $10.14/kg (2007$).

[d] Assume that the cellulase enzyme is Youtell #6 purchased from Chinese enzyme market at 13 Chinese Yuan (RMB)/kg enzyme with the protein content of 9% (Fang et al., 2013), equivalent to 13/6.2 (the present exchange rate of dollar to RMB)/0.09 = $23.30/kg (2013$).

electricity generation by lignin residue and methane combustion. 1.51 tons of lignin residue (50% of pure lignin content, w/w) per metric ton cellulosic ethanol is produced in DryPB process according to the Fig. 2. The correspondingly electricity generation from combustion of lignin residue and biogas is in the range of 7,121–8180 kWh from the present DryPB process (Liu and Bao, 2017). Therefore, the high electricity consumption of biorefining process is easily compensated with a high surplus of extra electricity.

3.4. Case analysis

The minimum wastewater generation and energy consumption of DryPB is achieved under the high bioconversion efficiency. Assuming that the bioconversion efficiency (ethanol titer, yield and cellulase dosage) in DryPB applies to other biorefining processes (DAP, AFEX, DMR and SE), the re-calculated wastewater generation reduced by 8–58%, and the electricity consumption reduced by 6–27%, a (Table 2). Certainly, the same bioconversion efficiency assumption practically does not stand due to the differences of pretreatment severity and inhibitor removal efficiency. We further assume that the solids state biodetoxification in DryPB is applied to other biorefining processes and the inhibitors are quickly and completely removed from the pretreated lignocellulose feedstock and xylose is well maintained. In such a scenario, the bioconversion efficiency elevation could be elevated, or the wastewater generation and energy consumption could be reduced (Table 2). For DMR and SE, the wastewater generation and steam consumption of could be reduced by 53–92% and 9–31%, respectively, if water washing detoxification is replaced by biodetoxification and the bioconversion maintains the same with DryPB. For AFEX, no detoxification step is involved but the inhibitors certainly exist and negatively affect the bioconversion efficiency with the reported maximum ethanol titer of 40 g/L (Uppugundla et al., 2014; Kim and Dale, 2015). DAP uses the ammonia conditioning for detoxification but results in the generation of high ammonia concentrated wastewater. The application of biodetoxification to AFEX and DAP could deliver an important solution for elevation of bioconversion efficiency close to DryPB.

Cellulase is the second largest item of cellulosic ethanol production besides the feedstock, although a high cost reduction has been achieved in the past decade (Humbird et al., 2011). There are two options of cellulase enzyme supply, one is the cellulase purchase from outside makers, and the other is on on-site production of cellulase enzyme within the biorefinery plant range. The present MESP calculation is based on the on-site cellulase production with the cellulase cost only $4.74/kg enzyme protein with $0.19/gal of ethanol in which the expensive purification, storage and transportation are not taken into account. If cellulase is supplied by the purchase from outside makers, however, the cellulase cost may increase to $5.07/kg enzyme protein at the minimum (same protein price to soybean protein) with MESP of $1.83/gal of ethanol (Kazi et al., 2010), or even to $23.30/kg enzyme protein at the current market price with MESP of $2.64/gal of ethanol (Fang et al., 2013) (Table 3). The impact of enzyme supply mode and price on MESP is remarkable. Therefore, the on-site cellulase production in the biorefinery plant is not only the key factor of cost reduction, but also the way to reduce uncertainty of cellulase enzyme supply.

Although the cellulosic ethanol production by DryPB technology is very close to corn ethanol production by dry mill technology in both technical and economical viewpoints, there are still significant differences between the two pathways: (1) capital investment. The major byproduct of cellulosic ethanol production is the large quantity of lignin residue used as combustion fuel to generate electricity, while corn ethanol produces the small quantity of DDGS used as animal feed. Construction of electricity generation plant requires much higher capital investment and operation cost than the DDGS processing step. (2) Feedstock collection chain. A relatively long and costly logistic chain is required to move the low density agricultural residue feedstock from the agriculture field to the gate of the biorefinery plant, comparing to the relatively short and available logistic of the high density corn grain feedstock. (3) Enzyme supply mode. Cellulose hydrolysis requires approximately two orders of magnitude greater cellulase enzyme than the alpha-amylase and glucoamylase in corn starch hydrolysis. Therefore, the purchase mode from the outside makers in amylase enzyme supply is not valid for cellulase enzyme. The on-site cellulase enzyme production unit should be included in the biorefinery plant, which increases the capital investment and operation cost. (4) Processing technology maturity. There are only few commercial plants under irregular operations in Europe and USA, certainly the technology maturity is below the advanced corn ethanol processing technology.

4. Conclusion

A new biorefining technology DryPB by combining a series of “dry” biorefining processes, demonstrates the significant advantages on minimizing energy consumption and wastewater generation while the high conversion efficiency is maintained. Ethanol titer, wastewater generation, steam consumption and overall cost of cellulosic ethanol by DryPB are close to that of corn ethanol. The disadvantage of high electricity consumption is easy compensated by lignin residue combustion. Cellulosic ethanol by the current DryPB process provides the highly competing potential to corn ethanol from either technical or economic viewpoint, under the proper capital investment, feedstock logistic system and on-site cellulase supply chain.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.biortech.2017.08.070.

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