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# Novel heat-integrated and intensified biorefinery process for cellulosic ethanol production from lignocellulosic biomass

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## ABSTRACT

Biofuels have the most potential as an alternative to fossil fuels and overcoming global warming, which has become one of the most serious environmental issues over the past few decades. As the world confronts food shortages due to an increase in world population, the development of biofuels from inedible lignocellulosic feedstock may be more sustainable in the long term. Inspired by the NREL conventional process, this paper proposes a novel heat-integrated and intensified biorefinery design for cellulosic ethanol production from lignocellulosic biomass. For the preconcentration section, heat pump assisted distillation and double-effect heat integration were evaluated, while a combination of heat-integrated technique and intensified technique, extractive dividing wall column (EDWC), was applied to enhance the process energy and cost efficiency for the purification section. A biosolvent, glycerol, which can be produced from biodiesel production, was used as the extracting solvent in an EDWC to obtain a high degree of integration in a biorefinery context. All configuration alternatives were simulated rigorously using Aspen Plus were based on the energy requirements, total annual costs (TAC), and total carbon dioxide emissions (TCE). In addition, the structure of the EDWC was optimized using the reliable response surface method, which was carried out using Minitab statistical software. The simulation results showed that the proposed heat-integrated and intensified process can save up to 47.6% and 56.9% of the TAC and TCE for the purification section, respectively, compared to the conventional purification process.

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## 1. Introduction

Over the past few decades, the production of biofuels, which are more environmentally sustainable fuels, has stimulated intense interest as a substitute for fossil fuels. Among the biofuels, bioethanol is one of the most promising candidates and has attracted considerable attention. The first generation bioethanol was derived from sugar or starch produced by food crops, which are considered critical to feeding the world's increasing population; therefore, its production may create a food-versus-fuel scenario. On the other hand, the abundant and relatively low-cost lignocellulosic materials make them attractive as a feedstock for the production of sufficient amounts of bioethanol from renewable resources at a reasonable cost [1]. The production of cellulosic ethanol (CE) from lignocellulose, which was estimated to produce approximately 86% less CO<sub>2</sub> emissions than oil sources [2], might be more sustainable.

Basically, the production of CE has three main steps: pretreatment of raw materials, bioethanol fermentation, and separation.

Among them, the energy-intensive separation process comprises a major proportion of the overall production cost [3]. Consequently, improvements of the separation process may have a dominant effect on the bioethanol market price. The function of the separation process is to produce a commercially pure CE product from a fermentation broth composed of 5–12 wt.% of CE with the balance being many other components. Note that most studies of the bioethanol separation process assumed that the feed from the fermentation broth was comprised of only bioethanol and water; hence, the separation of a homogeneous azeotropic mixture of ethanol and water (95.63 wt.% ethanol) was investigated. Obviously, this assumed feed leads to a simpler process but so far from the real biomass-based feed composition. In fact, breaking down the cellulose-hemicellulose-lignin structure of lignocellulosic biomass leads to the presence of more compounds in the output of the fermentation step, resulting in a more complex separation step. Therefore, a study of the CE separation process from lignocellulosic biomass should consider a real fermentation broth feed, which usually consists of water, CE, gases, soluble solids, and insoluble solids to reliably reflect the lignocellulose-based process.

Owing to the presence of an azeotrope between ethanol and water, the separation process is usually divided two parts:

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preconcentration part that concentrates ethanol close to the azeotropic composition and a dehydration part that produces anhydrous ethanol with commercial purity. Several technologies for the preconcentration process have recently been investigated, such as cyclic distillation [4] and membrane separation [5]. Although these technologies appear promising because of their high energy efficiency and low investment cost, they have not been exploited on an industrial scale. Liquid–liquid extraction [6] may be effective from an energetic point of view; however, an additional solvent regeneration part is needed. Typically, distillation is the most widely used preconcentration process on an industrial scale, because of its large capacity, economic benefit, and easy implementation.

The dehydration part that concentrates the CE to a high purity above the azeotropic composition requires non-conventional and more complex techniques. A number of dehydration alternatives have been reported: adsorption, extractive distillation (ED), azeotropic distillation (AD), pervaporation, vapor permeation, and pressure swing distillation [3,7–9]. Among them, the capacity of adsorption, pervaporation, and vapor permeation have reached their limits and are cost-intensive on an industrial scale [10]. Meanwhile, ED is more energy-effective than AD [3] and is commonly used in industry for the production of anhydrous bioethanol. ED uses an additional non-volatile solvent to alter the relative volatility of the components, resulting in the possible separation of an azeotropic mixture by distillation. The normal structure of ED consists of two columns, in which the first separates one pure component while the second recovers the solvent with the highest boiling temperature.

The selection of a suitable solvent plays an important role in ED design and optimization. Currently, ethylene glycol (EG) is the most common solvent applied in industry to separate ethanol–water mixtures [11]. On the other hand, it has serious environmental issues, such as fatal intoxication by ED poisoning [12]; and can also cause some health effects, e.g., central nervous system depression, cardiopulmonary effects, and renal damage [13]. As a result, a green solvent is needed as a substitute for EG. Bioglycerol, which is available as a byproduct in biodiesel production is a promising solvent alternative. The use of bioglycerol as an extractive solvent can achieve a high degree of integration in a biorefinery context, leading to a more sustainable ED process in the long term.

In recent years, several commercial scale processes for CE production have been constructed in the US, Brazil, and Europe. The first commercial scale CE plant producing 75 million liters of CE per year, which is currently the largest advanced biofuels refinery in the world, was opened in 2013 at Crescentino, Italy. Another plant using Beta Renewables technology began production in 2014 at Alagoas, Brazil. In the US, a 30 MMgy CE plant in Imperial Valley, California will also begin production in 2016. In addition, a number of demonstration plants and pilot plants are also under development. Nevertheless, due to the complexities of lignocellulose feedstock and biorefinery technology, more intense research and advanced designs are needed to make CE more cost-competitive with petroleum-based fuels and corn ethanol.

An innovative solution to overcome the disadvantages of an energy-intensive separation process is to apply advanced heat-integrated and intensified techniques, such as heat pump assisted distillation, double-effect heat integration, thermally coupled distillation, and dividing wall columns (DWC) [14–17]. Remarkably, DWC is one of the best examples of a proven intensified technique in distillation because it can reduce the operating and investment costs significantly while also achieving a great reduction of CO<sub>2</sub> emissions [15–17]. Recent studies reported the use of a DWC to enhance the bioethanol dehydration process [7,9], but they did not consider the most energy-intensive step, the preconcentration process. To the best of the authors' knowledge, none of the studies

used advanced process intensification and integration techniques for the whole bioethanol separation process from an actual fermentation broth, particularly with those processes using lignocellulosic feedstock as the raw material.

This paper proposes a novel heat-integrated and intensified biorefinery design for CE production from actual lignocellulosic biomass. For the preconcentration section, heat pump assisted distillation and double-effect heat integration were evaluated. For the dehydration section, in addition to heat integration, process intensification, which is an extractive dividing wall column (EDWC), was applied to enhance the energy and cost efficiency of the purification process. Furthermore, a biosolvent, bioglycerol, which may be produced from biodiesel production was used as an extracting solvent in EDWC to obtain a high degree of integration in a biorefinery context. The structure of the EDWC was optimized using the reliable response surface method (RSM), which was carried out in Minitab statistic software. All configuration alternatives were simulated rigorously using Aspen Plus and evaluated based on the energy requirements, total annual costs (TAC), and total carbon dioxide emissions (TCE). Moreover, the contribution of proposed process on the absolute production cost for CE that can be used to assess its competitiveness and market potential with corn ethanol and petroleum-based fuels was explored.

## 2. Methodologies

### 2.1. Design

In this study, inspired from the CE production process developed at NREL (National Renewable Energy Laboratory in the US) [8], several alternatives for the separation process of CE were proposed and simulated using Aspen Plus V8.8. The lignocellulosic biomass type used in this process was corn stover, which is the most abundant agriculture residue in the US. This feedstock was comprised of 17.9 wt.% soluble material (ethanol, sucrose, extractable inorganics, and other water extractable material), 31.9 wt.% glucan, 18.9 wt.% xylan, 13.3 wt.% lignin, and the balance being other components [18]. Fig. 1 illustrates the cost contribution details from each process area (per gallon CE) in the NREL process [8]. The distillation and solids recovery section, which is the motivation of this study, accounts for 5.6% of the CE selling price (\$2.14/gallon in 2007\$). In this purification process, the feed mixture with detailed composition listed in Table 1 is the fermentation broth obtained from the CE fermentation process. This complex broth stream contains a large number of compounds in three phases, resulting in a more complicated separation process.

Fig. 2 presents a systematic procedure to improve the CE purification process. First, the CE base case was designed and optimized based on the NREL reference case [8]. In particular, the pre-concentration part was kept same as the NREL case while the dehydration part was designed using extractive distillation with bioglycerol solvent. After achieving the optimal base case, the CE process was then explored for heat integration opportunities. Two common heat integration techniques such as heat pump and double-effect were applied and evaluated in detail. In the next step, process intensification techniques were assessed to improve process performance further. Two columns in the traditional ED configuration were combined into one compact column, EDWC. Subsequently, the feasibility of combining both process intensification and heat integration techniques was carried to maximize the process efficiency. In addition to energy-saving comparison, all alternatives were also compared in terms of process economics and environmental impact. Through this framework, a promising process for CE purification from actual fermentation broth was proposed with detailed design and operating condition.

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