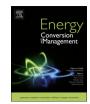
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## Analysis of process intensification and performance assessment for fermentative continuous production of bioethanol in a multi-staged membrane-integrated bioreactor system



Parimal Pal<sup>a,\*</sup>, Ramesh Kumar<sup>b</sup>, Alak Kumar Ghosh<sup>b</sup>

<sup>a</sup> Environment and Membrane Technology Laboratory, Department of Chemical Engineering, National Institute of Technology Durgapur, 713209, India
<sup>b</sup> Department of Chemistry, The University of Burdwan, 713104, India

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#### ABSTRACT

The work analyses process intensification in production of fermentative biofuel (bioethanol) in a multi-staged membrane integrated bioreactor system from sugarcane juice (SCJ) using Saccharomyces cerevisiae (NCIM 3205). Membrane based clarification, sterilization and concentration of sugar cane juice could replace several energyintensive and polluting steps of conventional production schemes. Integration of traditional bioreactor with membrane-based devices for downstream purification of bioethanol enabled the system for recycling of cells and residual sugars to the fermentation unit through largely fouling free cross flow microfiltration and nanofiltration membrane modules. The new process design is characterized by high yield (0.48 g/g), productivity (14.6 g/L/h) and concentration capability (97.6 g/L) using concentrated SCJ of 22 wt% under high cell density in a flexible, compact, eco-friendly and modular plant. The final membrane distillation stage concentrated ~98% pure ethanol in a solar-driven direct contact membrane distillation configuration using PTFE/PET hydrophobic membrane with enhanced flux with vis-à-vis the existing processes. A rectangular flat sheet cross flow membrane module in counter-current flow mode of hot and cold streams was employed. The membrane-based system achieves quite high levels of process intensification essential to sustainable operation at industrial scale. The achieved process intensification in the new system has been analysed vis-a-vis the existing conventional systems in terms of eco-friendliness, flexibility, cost of equipment and production, consumption of energy and material, profitability, E-factor, atom efficiency and business sustainability.

#### 1. Introduction

Among different available biofuels, bioethanol blended gasoline is considered as one of the most eco-friendly transport fuels. From the current consumption pattern of fossil fuel, International Energy Organization (IEO) forecasts an enhancement of 56% in energy consumption over the 30 year period of 2010–2040. More than 80% of the consumed energy is supplied by fossil fuels leading to emission of greenhouse gases (GHG) which in turn results in global warming [1]. Excessive use of fossil fuels is leading to total exhaustion of oil reserves within the next 50 years. Thus the issues of energy security and environmental safety have turned the awareness of the current world to explore alternate and eco-friendly energy source like wind, geothermal wind, water and biomass. Among these possibilities, ethanol derived from renewable sources like biomass is one of the most promising sustainable energy as it can be mixed with gasoline for transport fuel with reduced harmful emissions like that of CO and hydrocarbons [2]. Several countries spreading over Asia, Europe and America have shown their commitment towards bioethanol production programs to reduce dependence on fossil fuels. For example, ethanol blending in petrol is now around 10% in India and USA whereas it is 27% in Brazil.

A paradigm shift in production strategy is now imminent in generation of energy and in chemical, pharmaceutical and allied industries process industries to switch over from conventional energy-intensive and largely polluting production schemes to green production regime [3]. Such green production strategy encompassing substantially more efficient, energy-saving, compact and flexible production systems for a given performance level is often termed as process intensification (PI). In this era of emaciated profit margin, PI may be the right strategy towards business sustainability with the promise of reduced material and energy consumption in compact, flexible and modular plant designs. The expositions of PI is on intensifying the traditional processes to smaller and cleaner process plants with reduction in equipment size, cost, energy, manpower and unit operations especially in reaction

\* Corresponding author. *E-mail addresses*: parimalpal2000@yahoo.com, ppal.nitdgp@gmail.com, parimal.pal@che.nitdgp.ac.in (P. Pal).

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engineering [4]. In order to find the optimal conditions to get the maximum profit, decisions are often taken on re-design of equipment or replacement of some processes in scientific, industrial or commercial activities [5]. Membrane based downstream separation-purification is emerging as an effective approach in chemical production processes where membranes are being used as membrane reactor, membrane distillation unit, membrane contactors, membrane adsorption and membrane crystallizer units as parts of the processes [6]. The same approach is well extendable to biodiesel production also in view of the separation-purification steps involved. Ethanol has some excellent properties such as its antiseptic nature, low toxicity, low boiling point, high octane number and miscibility with water. These properties permit its use as transport fuel in addition to use in medicine (antiseptic, antibacterial sanitizer and drugs) and in chemical industries as solvent for producing perfumes, varnishes and flavoring agents [7].

Feedstock for bioethanol production should be cheaper and easily available in sufficient quantity in nature and renewable like sugarcane, corn maize, cassava, Mahua flower and agro-industrial wastes. Corn is the prime substrate used for ethanol production in US whereas sugarcane and molasses are used in Brazil and India. Mahua flower (*Madhuca latifolia* L.) has a droopy fleshy off containing 66–72% sugars which is traditionally used by tribal communities to produce alcoholic beverages using yeast called as "mahuli" [8]. Two microorganisms viz. yeast (*Saccharomyces cerevisiae*) and bacteria (*Zymomonas mobilis*) are mostly used for bioethanol fermentation. Moreover, Behera et al. [9], found that *Saccharomyces cerevisiae* could produce 149 g/kg of ethanol against 123 g/kg by the *Zymomonas mobilis* on 96 h of fermentation. In this study, fermentable sugar extracted from sugarcane was used in bioethanol production using fermentation with yeast (*Saccharomyces cerevisiae*).

The products bioethanol and water form an azeotropic mixture in the ratio of 95:5 during conventional distillation. Economical production of bioethanol needs to overcome the issues like high energy consumption and azeotropism where in the conventional refining process,  $\sim$  40% of the total energy is consumed in dehydration of ethanol after fermentation [10]. In conventional distillation of ethanol from water, energy required to concentrate from 92.5% to 95.6% increases exponentially due to ethanol-water azeotropic formation [11]. However, using molecular sieve with zeolite beads techniques, Al-Aheh et al. [11] increased the ethanol concentration from 91 wt% to 99 wt%. Glycerol easily available, low cost and can break the azeotropism during extractive distillation thereby facilitating ethanol dehydration [12]. During dehydration, drying chemicals like benzene is re-circulated continuously in small quantity as drying agent. In advanced approach of extractive distillation, a dividing wall column combines a solvent recovery column with other extractive columns for dehydration of ethanol using ethylene glycol and n-pentane. 99.8% of concentrated bioethanol is achieved in this approach [13,14]. After dehydration, denaturation needs to be done to meet the criteria for addition in gasoline. All these steps are energy-intensive where 1 L of ethanol production needs 7.4 kg of steam generation to meet the energy demands [15].

In addition to that, in conventional batch type fermentation process, the product-inhibition is observed when ethanol concentration reaches 12% by volume due to hindrance in the specific cell growth rate of the microorganisms [16]. To overcome these problems, PI with multi-staged membrane integrated bioreactor system for continuous bioe-thanol production is required which has the potential of completely replacing the conventional production system. Such a membrane-based plant can overwhelmingly simplify and replace several conventional downstream production steps like centrifugation, distillation, acid-ification, neutralization, adsorption, ion-exchange and dehydration. These high energy consuming operational units may be eliminated on integration of membrane based downstream purification steps with fermentation units for product recovery. The target molecules may be fractionated in its purest form using tailor-made membrane systems.

This opens a new route for the separation and purification of different products in pure form [17]. A number of advantages emanate from such membrane based processes such as reduction of operational units and energy consumption with enhanced yield, productivity and high cell density without any substrate-product inhibition. Pressure, chemical or thermal-driven membrane systems like microfiltration (MF), ultra-filtration (UF), nanofiltration (NF), pervaporation (PV) and membrane distillation (MD) are used in bio-refining processes [18]. The potential of application of membrane systems during downstream purification of bioethanol after fermentation of enzymatically pretreated renewable lignocellulosic material as carbon source has been reviewed [19].

Membrane distillation may be successfully applied for final stage of downstream purification and recovery of bioethanol using microporous hydrophobic membranes such as polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF) and polypropylene [20]. Woldemariam et al. [21] found that membrane based distillation could be a competitive technology for replacement of distillation column for ethanol recovery using waste heat from industrial-scale bioethanol plant with 0.87 M US \$ yearly saving. Some authors have used membrane distillation immediately after fermentation to recover the bioethanol but with significant deterioration of the membrane permeability, selectivity and its efficiency due to presence of high biomass and sugar concentration [22,7]. Ethanol recovery using direct contact membrane distillation (DCMD) was integrated with BIOTRON reactor for ethanol production from lactose solution by Gryta et al. [16] and they observed that partial or full wetting of membrane as confirmed by FT-IR analysis due to infiltration, adsorption of yeast cells and/or sugar molecules. This may eventually limit the industrial scale application. However, when clear fermentation broth obtained after microfiltration and nanofiltration is used as feed for DCMD in proper module design, membrane fouling and wettability problems can be reduced significantly during membrane distillation to recover ethanol. Thus, an integration of cross-flow membrane module for MF and NF after conventional fermentation has been done to recycle the cell biomass and unconverted sugar molecules and finally recover the concentrated bioethanol using solar driven DCMD to overcome the major difficulty in sustainable operation to achieve the PI. In present study, integration of bioreactor with multi-staged membrane system for the production, separation, purification and concentration of bioethanol from a cheap and easily available carbon source was investigated with analysis of culminating PI leading to the sustainability in the backdrop of absence of a similar study in the literature.

#### 2. Recent trends in ethanol production

Bioethanol from sugar-based raw material through fermentation is referred as "first generation" bioethanol whereas such bioethanol from lignocellulose as raw material is commonly known as "second generation" [23]. Now in third generation, algal biomass is under investigation for ethanol production [24]. However, the pretreatment processes of 2nd generation and 3rd generation based biomass involve many technical and economic challenges as compared to 1st generation sugar based feedstock [25]. Initially ethanol was used to produce by hydration of ethylene - a petrochemical by-product, at high pressure and temperature in presence of catalysts phosphoric acid adsorbed in silica, used as industrial feedstock or solvent. Moreover, ethanol used for medicine, alcoholic beverages and fuel produced by fermentation using yeast and different carbohydrate sources [26,27]. Distillation is the most commonly used method for the downstream purification and dehydration of bioethanol after fermentation. However, there is no clear technical or commercial advantage in both biochemical and thermochemical pathways for ethanol productions till date. For the ethanol production from biomass through fermentation, much improvements needs to be done, like selection and characterization of feedstock, pretreatment cost reduction by improving efficiency of enzyme and recycling/reusing and improving overall processes integration [28].

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