Development of an advanced enzyme reusable saccharification process of waste paper pulp sludge materiel through membrane bioreactor system: a concept towards green solid waste management practices for PPS material

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Abstract

Amidst worldwide concern for solid waste management practices of PPS material mainly includes landfilling, incineration and burning which leads to wastage of valuable bio-energy resources and raises challenges towards environmental safety. Exploiting PPS as a source of cellulose derived energy, value added chemicals like fermentable sugars are most emerging processes which got substantial attention to mitigate such environmental problem. Enzymatic saccharification for synthesis of fermentable sugars has distinct advantage as the process occurs at mild environmental condition with maximum yield due to reaction specificity.

However, major obstacles to practical realization of large scale enzymatic hydrolysis include enzyme cost, energy consumption during high solid loading process, product inhibition and enzyme recovery. Selection of ideal pretreatment, adaptation of suitable solid loading, utilization of native crude enzyme or low cost commercial enzyme, periodic product removal, recovery and reutilizing enzymes with suitable bioreactor design are most sustainable approaches to improve the efficiency and economy of the process. A suitable membrane bioreactor reactor system can ensure enhanced production and separation of simple sugars after saccharification process of PPS material in order to avoid product inhibition problem while confirming enzyme reusability. Even surface modification of conventional polymeric membranes can further improve the separation efficiency of enzymes.

Keywords: Paper sludge, Enzymatic saccharification, membrane reactor, surface modification.

Introduction

Paper has become one among the most used utilities in the world today. This has caused excessive demand for paper and paper related products around the world. Paper production demands the cutting down of a large number of trees. This has not only led to widespread deforestation but also environmental pollution since paper mills are one of the main sources for air, water and soil pollution. Pulp and paper sludge (PPS) is the main organic solid by-product generated during wood pulping and paper making operations. Landfilling, incineration and burning are the common solid waste management practices with secondary PPS material and such procedures mainly leads to wastage of valuable bio-energy resources, significant cost increasing during paper production and raises challenges towards energy security, environmental safety by polluting environment. Discarded paper sludge has become one of the major components for filling up landfill sites. The total amount of municipal waste produced in the United States of America was 262.4 million tons, out of which waste from paper and cardboard waste accounted for nearly 25.9% of the total amount in the year 2015.

Unfortunately, the recycling of paper also leads to widespread contamination of natural sources due to the de-linking process that is being carried out. Indian paper industry is poised to grow and touch 25 million tones at 2019-20 from 20.37 million tones at 2017-18 according to the Associated Chambers of Commerce and Industry of India. The paper pulp industry is the third largest industry which contributes to air, water, soil pollution in countries like Canada and US and releases 100 million kg of toxic pollutants each year. Paper industry consumes more water to produce a ton of product than any other industry in the world. Presently, many countries all over the world follows legislative trend to restrict types and amount of material permitted for landfilling[1]. The major environmental concern with PPS landfilling includes ground water contamination from disposal practices, soil degradation, methane emissions and increased risk of greenhouse gas (GHG) emissions[2]. To eliminate the negative environmental impact like climate change generated from landfilling of PPS material, several solid waste management practices have brought under consideration.

The novel property of the paper sludge is that during the paper pulping process, maximum amount of lignin is removed from the paper sludge material. As a result, it does not necessities further expensive pretreatment strategies for separation of lignin and can be used directly for production of value added products. Recent trends like the usage of microbes to degrade the paper sludge and convert them into compost are not that effective and it is considered to be a costly approach. Hence developing and undeveloped countries cannot undertake such costly approach. The only solution to this imminent problem is to develop methods to convert the sludge into value-added products using technologies that do not cost much. Exploiting primary PPS as a source of cellulose derived energy, value added chemicals like bio-ethanol, biogas, bio-hydrogen and bio-butanol are the most emerging processes which got substantial attention to control future energy insecurity problem, caused by the disposal of such material. Enzymatic saccharification process is the most important intermediate stage which determines the overall efficiency of each such process.

Hence it became important to strengthen the enzymatic saccharification process for production of enhanced amount of fermentable simple sugars which can lead to several valuable fermentable end products. Among different saccharification strategies, high solid loading fed-batch operations are found to be more effective compared to the batch processes. The main obstacle for conventional enzymatic hydrolysis is the high cost of enzyme, product inhibition effects and lack of understanding of cellulase kinetics on lignocellulosic substrates. One way to overcome these problems are to operate the enzymatic hydrolysis using high insoluble solid loading and simultaneous removal of enzyme and end product.

However, the saccharification reaction at high insoluble solid loading conditions will have to encounter the problems of increased viscosity, the higher energy requirement for mixing, shear inactivation of cellulases and poor heat transfer due to rheological properties of dense suspension[3]. Interestingly through fed-batch enzymatic hydrolysis, such problems could be avoided by adding the substrate and or enzymes gradually to maintain a lower viscosity level[4]. The fed-batch enzymatic saccharification process has several other economic advantages over conventional batch process such as lower capital cost due to reduced volume, lower operating costs and lower down-stream processing cost due to higher product concentration[4].

In the recent years, membrane based hybrid reactors received considerable attention for manifestation of multiple tasks in a single stage through highly selective and efficient transport properties. More specifically, such reactor system can play effective role in improved enzymatic saccharification process by continuous removal of inhibitory end products, continuous separation and reutilization of enzymes. Over the last two decades application of both submerged and side stream membrane reactor system have mainly remain confined in the field of municipal and industrial waste water management and less investigated for enzymatic saccharification process. With comparison to side loop membrane reactor system, submerged or immersed membrane bioreactor ensures share sensitive enzyme hydrolysis system with lower energy consumption, sustainable processing, simpler operation including cleaning and relatively easy scale-up provisions. Application of PES or PES-based membranes ensures long time separation performances of enzymes due to their comparative stability but hydrophobic nature makes them more affected by fouling problem. To address the issue, some robust and extensive surface modification methods were attempted by large number of researchers. Among such methods, surface coating techniques involves the use of chemicals under risky and hazardous conditions[5] whereas plasma and UV irradiation methods are not suitable for largescale industrial applications[6,7].

Polymeric blending with hydrophilic polymers, such as polyvinylpyrrolidone (PVP) or polyethyleneglycol (PEG) is considered most effective technique as such non-solvent additives are acting as efficient pore forming agent which helps to maintain maximum rejection of solute molecules without affecting permeate flux. To maintain fouling free uniform operation of membranes, solid residues after saccharification will be removed and such strategy will be even useful for reutilization of non-soluble solid adsorbed enzymes. In the present review, effectiveness of such promising, compact and less energy-intensive membrane based saccharification scheme of PPS material for production of valuable fermentable sugars is presented. To our knowledge, such concept of membrane based efficient enzymatic saccharification strategy of PPS material is absent in the existing literature.

Pulp and Paper sludge material (PPS): Paper sludge is a solid by-product of paper making operations which is considered as a waste product. This paper sludge is a lignocellulosic waste material which mainly consists of cellulose and lignin. Fibres present in paper can be recycled nearly five times depending on the type of paper which is being produced. Wastes generated from paper and pulp industry are basically classified into four categories which includes (i) rejects in terms of lumps of fiber, metals, glass and sizing agents (ii) deinking sludge mainly containing short fibers, inks and other additives (iii) primary sludge and (iv) secondary sludge[8]. Total process involves 300 to 2600 m^3ofwater for treating per ton of printing and writing paper and it approximately generates 50 kg of primary sludge material in terms of dry solid[8].

Estimated annual production of paper and paperboard throughout the world is 400 million tons[2]and it is predicated to be increased at 550 million tons by 2050 which could enhance the production of PPS by 48-86% with comparison to its present rate of formation[9]. Mainly process and wastewater treatment technology decides the amount of paper sludge production. Almost around 40–50 kg dry sludge with composition primary (70%) and secondary (30%) sludge is generated from per ton of paper manufacturing process[10]. Paper mill sludge mainly secondary sludge material is enriched with organic contents, important plant nutrients including N, P, potassium (K), Ca and Mg, along with other inorganic ions but at the same time it contains inorganics like clay, ash, fillers, etc. Production of paper sludge from paper manufacturing unit is presented in figure 1.

Exploiting primary PPS as a source of cellulose derived energy, value added chemicals are the most emerging processes which got substantial attention to control future energy insecurity problem and can balance the challenges generated through the disposal of PPS material. In terms of carbohydrate, PPS material consists of 27-47 % of cellulose, 4.84-14.2 % of hemicellulose and around 15.4 % of acid soluble, insoluble lignin[11, 12]. Additionally, PPS provides some distinctive advantages for production of value added chemicals compared to other lignocellulosic material. It includes (i) PPS materials are relatively abundant and economically compatible as feedstock cost is comparatively low (ii) they contain comparatively high levels of carbohydrate (iii) reduction in cost or energy required for energy-intensive pretreatment process as extensive pulping process effectively de-lignifies the initial biomass and removes significant amount of poorly fermentable hemicellulose materials[13,14].

Another distinct feature of such production process is that PPS biorefinary can be easily integrated with a preexisting infrastructure of paper mill operations[15].Due to the presence of high content conversable hemicellulose and cellulose and attractive features, PPS material is believed to be the most promising feed stock for production of value added chemicals like bio-ethanol, biogas, bio-hydrogen and bio-butanol through fermentative route[8,11]. In the form of main impurity, amount of clay present in paper sludge material is about 10.5 % but it does not show any inhibitory effect in fermentative production processes and can be removed completely by subsequent washing of the material. Among all, PPS material is mostly exploited for bio-ethanol production[11,16,17] but still now not much distinctive research efforts have been made for development of most simplified, effective and economic technology which mainly supports in production of concentrated form of fermentable simple sugars through high solid loading saccharification process of paper sludge material. The successes of the process may support in the development of various value added products through fermentative production routes.

Enzymatic Saccharification: For enhanced production of fermentable simple sugars, high solid loading enzymatic saccharification strategy is considered as ideal approach. Researchers like Yang et al[18], Liu et al[19], Gao et al[20] and Sugiharto et al[21] analyzed the impact of high solid fed batch enzymatic scarification process for enhanced production of simple sugars in their different case studies. Liu et al[19] and Yang et al[18] achieved maximum solid loading of steam treated corn stover biomass up to 30 % (w/w) and 12 % (w/w) in fed –batch enzymatic hydrolysis process and steam treatment was identified as best routes for maximum separation of hemicellulose. Gao et al[20] clearly justified how high power consumption, stirring problem, mass transfer difficulties in high solid batch saccharification process from sugarcane bagasse can be overcome by fed-batch saccharification process. Liu et al[19] separately analyzed viscosity changes with processing time in batch saccharification process with respect to different substrate concentration in order to understand the suitable time for substrate feeding in fed-batch saccharification mode. But such solid loading saccharification strategies are ignored for fermentable simple sugar production from PPS material.

It was identified that saccharification step as most critical bottleneck to achieve maximum hydrolysis efficiency and the major influencing factor are:
(1) identification of kinetically active cellulase enzyme with better access to substrate
(2) sustaining with maximum activity of the enzyme under different acid-alkaline pretreatment condition or shear sensitive condition. Akhtar etal.[22] more focused on the development of cost-effective efficient pre-treatment techniques to achieve maximum separation of lignin and maximum hydrolysis efficiency.

Research efforts related to the high solid fed-batch enzymatic saccharification, identification of different additives for influencing enhanced production of simple sugars are lacking in national scenario specifically from paper sludge material.

Membrane Bioreactor for enzymatic saccharification: Several studies have highlighted ultrafiltration membrane reactor system is ideal for separation of enzymes while ensuring separate permeation, purification of simple sugars from saccharification process. Combination of ultrafiltration membrane separation unit with hydrolysis reactor enables separation of glucose and recovery of enzyme in a single integrated unit. Numerous enzymatic saccharification studies have been conducted while implanting external loop ultrafiltration membrane bioreactor for periodic or continuous removal of simple sugars. Such external loop membrane bioreactor requires circulation of mixed liquor containing cellulase enzyme through membrane module with high cross flow velocity. As a result, shear stress caused by the high velocity side stream configuration has a negative impact on enzymatic activity. On the other hand submerged membrane bioreactor is known for its low energy consumption as membrane is placed inside the membrane reactor or an external tank, hence it eliminates the requirement of external pumping. Configuration of external loop and submerged membrane bioreactor is represented in figure 2.

Even such submerged membrane reactor is considered more productive than immobilized enzyme catalytic reactor where enzyme is susceptible to conformational changes and leakage. Membrane cleaning is also difficult in such system.

Ylitervo et al[23] developed less energy consuming submerged membrane bioreactor system for fermentation of toxic lignocellulosic hydrolysate to ethanol. Flat sheet membrane panels were placed in an external chamber with provision of gas sparging for continuous separation and recirculation of biomass while avoiding membrane blocking and fouling. A new configuration of modified submerged membrane reactor was reported by Thaothy et al[24] for efficient enzymatic hydrolysis of cellulose with simultaneous glucose removal and enzyme recovery. Enzymatic saccharification was conducted in a sealed bag, made up of Nylon Monofilament Mesh (NMO) placed inside a 500 ml glass Pyrex beaker as a reactor which was also equipped with separate ultrafiltration device with PES membrane for continuous recovery of glucose. Conventionally, ultrafiltration membrane process with membrane molecular weight cutoff 10-100000 Da used for the recovery of cellulases after enzymatic hydrolysis but most of the existing studies[25-29] in this field adopted energy consuming, share sensitive external loop membrane reactor configuration. Most of the time, hydrophobic polymeric membranes like Polyether sulfone (PES) or Poly-sulfone membranes were implemented in those studies. Even different types of membrane modules were also investigated to analyze enzymatic saccharification efficiencies. Yang et al[30] analyzed the efficiency of the tubular reactor coupled with ultrafiltration membrane for continuous enzymatic hydrolysis of rice straw. Hwang et al[31] enabled 86% retention enzymes in an enzymatic hydrolysis process by using 10 k Dapoly sulfone membrane in a hollow fiber module. Cross flow membrane modules are comparatively better than the other configuration in terms of maintaining higher permeate flux at reduced fouling condition.

The principal impediments for the application of both submerged and side stream membrane bioreactor system are membrane fouling problem and affinity for adsorption of enzyme on the solid residue hindering its own recycling[32]. The second challenge can be easily mitigated by adopting variety of desorption method including the use of surfactant, alkali, urea and buffer with different pH[33]. Whereas several methods have been proposed to alleviate permeate flux reduction due to former problem such as increased aeration, relaxation of membranes and back wash, addition of chemical coagulants like alum, iron salts, modification of membrane surface with suitable material, selecting suitable hydrodynamic conditions while operating the membrane operation in subcritical flux, intermittent permeation and efficient module design[34,35].

It was clearly identified that submerged configurations are comparatively more advantageous in terms of lower energy consumption, overcoming high viscosity problem and flexibility of the membrane placement inside the external tank to control membrane fouling. Hawarietal.[34]investigated fouling suppression strategy in submerged membrane bioreactor system by using dielectrophoretic forces. The membrane bioreactor was used to treat biomass from waste water treatment plant in Bremen, Germany while implementing different electric field intensities (60-600 V) with variations in frequencies (50-1000 Hz). Specific approach by adding sponges into a conventional submerged anaerobic membrane bioreactor system was investigated by Liu et al. [36]to minimize membrane fouling and to improve treatment efficiency of synthetic waste water. Due to the potential advantages of membrane cleaning and replacement, transverse vibrational submerged membrane system was successfully used for separation and concentration of milk proteins in another case study. Hwang et al[31] pointed out that both filtration flux and protein rejection are the important issues to increase protein recovery and submerged membrane bioreactor is ideal to achieve it with reduced fouling.

In a similar case study, Malmalietal.[37]highlighted the potential of submerged membrane bioreactor system for economic hydrolysis of cellulosic material. Submerged polyethersulfonesulfone microfiltration membrane (0.65 µm) was used to withdraw glucose solution at the pressure slightly lower than atmospheric pressure. Rate of glucose production was determined by optimization of mixing, rector holding time, time of feeding and finally it ensures operating flux of 75 L/m^{2}h. Hydrophobic polymers such as polysulfone, polyethersulfone, polypropylene, polyethylene or polyvinylidene fluoride are mainly utilized for the formation of most of commercially available membranes. Among all such membranes, Polyethersulfone (PES) is considered as a “standard” material used for manufacturing UF membrane due to itsmechanical strength, thermal stability and chemical stability. PES membranes have been successfully applied in different biological applications like selective permeation, protein recognition/purification, controlled release, isolation of soya proteins etc.

Membrane surface modification: Hydrophobic polymers such as polysulfone, polyethersulfone, polypropylene, polyethylene or polyvinylidene fluoride are mainly utilized for the formation of most of commercially available membranes. Among all such membranes, Polyethersulfone (PES) is considered as a “standard” material used for manufacturing UF membrane due to itsmechanical strength, thermal stability and chemical stability. Even the membrane material has been widely used in food industry, including processed meat, nutritional beverages, infant formulas and dairy product replacements. But such hydrophobic polymeric membranes are prone to fouling[38] and hence application became limited for the separation of highly selective protein. Different techniques have been considered to enhance the hydrophilicity of such membranes through surface modification including coating techniques, blending method, plasma treatment, UV grafting and surface initiated atom transfer radical polymerization (SI-ATRP)[5,7,39].

In order to attach the hydrophilic polymer on the surface, coating techniques involves the use of chemicals under risky and hazardous conditions and hence this method is not considered environmentally friendly[5]. Similarly, for small-scale applications in chemistry laboratory, plasma and UV irradiation methods are considered very promising and effective technique but practically it is not suitable for largescale industrial applications[6]. Polymeric blending of functional groups has comparatively gained lot of attention to introduce advanced material properties in the membrane as it is easiest strategy to enhance the hydrophilicity of the membrane surface and it can be easily scaled up.

Several researchers attempted some robust and extensive method for surface modification of different membranes for the separation of proteins. Apparently, those methods are efficient in lab level but will be expensive for large-scale industrial applications. Amongst such modification approaches, the use of sulfonated poly(arylsulfone)s was identified as an efficient and attractive strategy to modify the surface charge, to create more hydrophilic surface for robust filtration membranes, to enhance the membrane permeability and to anticipate anti-fouling properties in the membrane[40]. Non-solvent induced phase separation method was implemented to develop ultrafiltration membranes from PSU and various types of sulfonated poly(arylsulfone)s. Dass et al.[41] fabricated PPSu (polyphenylsulfone) and SPPSu (Sulfonated polyphenylsulfone) /nanoparticle titanium oxide (TiO_2) hollow fiber membranes by phase inversion technique for developing antifouling properties and efficient separation of proteins.

Negatively charged organic–inorganic hybrid ultrafiltration membranes with adjustable charge density were fabricated by Kumar et al[42] for hybrid ultrafiltration based separation of protein in reduced fouling condition. In this study Sol-gel method was used to fabricate membrane by blending of water soluble poly vinyl alcohol and N-O-sulfonic acid benzyl chitosan (NSBC) in combination with tetraethyl orthosilicate (TEOS) silica precursor. In a similar case study, antifouling properties in the UF membranes was induced via grafting of positively charged poly(ethylenimine) (PEI) chains onto brominated tetra-methyl polyethersulfone (TM-PES) membranes for better separation of proteins like ferritin and bovine serum albumin (BSA)[43].

Polymeric blending with hydrophilic polymeric materials such as polyethylene glycol (PEG) or polyvinylpyrrolidone (PVP) are considered most simplest and widely used method to modify both flat-sheet and hollow fiber hydrophobic PES membrane with enhanced chemical and mechanical strength.

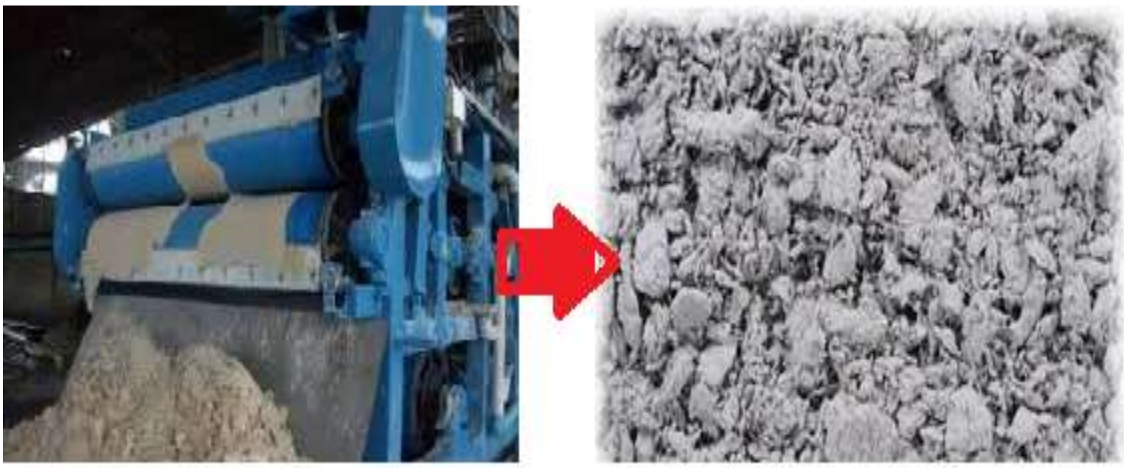

Figure 1: Production process of pulp and paper sludge material

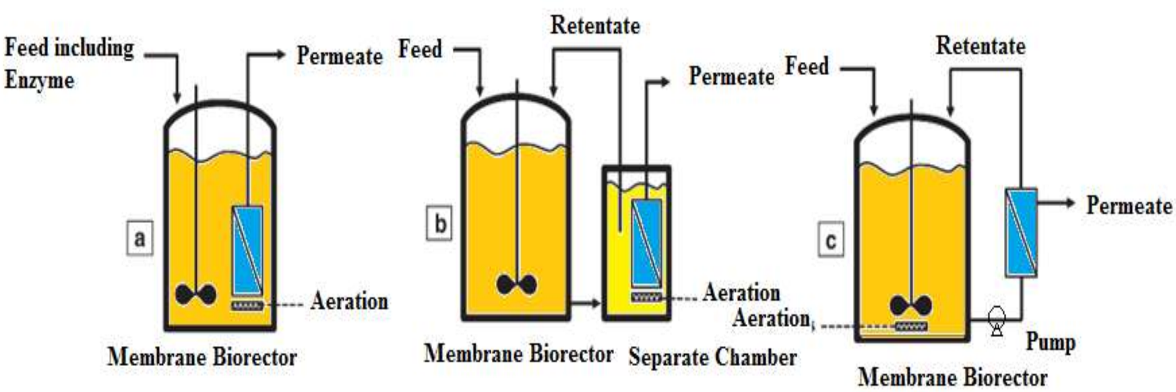

Figure 2: Configuration of different types of membrane reactor for enzymatic saccharification

Researchers like Liu et al[36], Wang et al[44] already implemented polymeric blending of efficient pore forming agent like polyethylene glycol (PEG) or polyvinylpyrrolidone (PVP) for improvement of PES membrane. The improved performances of those membranes were evaluated in terms of their flux enhancement, developing hydrophilicity, permeability and antifouling properties, improved separation efficiencies of proteins like bovine serum albumin, chicken egg albumin, lysozyme. Such high prospective surface modification technique of PES membrane is yet to be evaluated for separation and reusability of cellulolytic enzymes in improved saccharification process of lignocellulosic waste materials like PPS material.

Conclusion

Selection of paper pulp sludge material (PPS) among different lignocellulosic biomasses for biorefinery purposes is considered an ideal strategy as it offers several advantages. Still now PPS material is mainly utilized for bio-ethanol production and not much distinctive research efforts have been made towards enhanced production of fermentable sugars in purified form through improved enzymatic saccharification strategies. In this review, novel strategies like adaptation of high solid fed-batch hydrolysis, utilization of in house produced cellulase enzyme or low cost crude enzymes, enzyme recovery and reutilization of enzymes using ideal bioreactor configurations are projected to make overall saccharification process of PPS material economically viable.

Submerged ultrafiltration membrane bioreactor system is considered ideal for enzymatic hydrolysis of PPS material. Novel configuration of such bioreactor facilitates hybrid operations like separation, reutilization soluble enzyme to enhance saccharification efficiency and removal of reducible sugars in permeate side to avoid product inhibition effects. Polymeric blending approach with pore forming hydrophilic polymers like polyvinylpyrrolidone (PVP) or polyethyleneglycol (PEG) for membrane surface modification can contribute in better separation of proteins.

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