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Review

Integration of biological pre-treatment methods for increased energy recovery from paper and pulp biosludge

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ABSTRACT

The paper and pulp industry (PPI) produces high quantities of solid and liquid discharge and is regarded as the most polluting industry in the world causing adverse effects to environments and human beings. Hence changes in the way PPI sludge and waste materials are treated is urgently required. Nearly, 10 million tons of waste is generated per year, however PPI waste is enriched with many organic chemicalscontaining a high percentage of lignin, cellulose, and hemicellulose which can be used as valuable raw materials for the production of bioenergy and value-added chemicals. Pretreatment of complex lignocellulosic materials of PPI waste is difficult because of the cellulose crystallinity and lignin barrier. At present most of this waste is recycled in a conventional treatment approach through biological and chemical processes, incurring high cost and low returns. Henceefficient pretreatment techniques are required by which complete conversion of PPI waste is possible. Therefore, the present chapter provides the scope of integration of pretreatment methods through which bioenergy recovery is possible during the PPI waste treatment. Detailed information is presented on the various pre-treatment techniques (chemical, mechanical, enzymatic and biological) in order to increase the efficiency of PPI waste treatment and energy recovery from PPI waste. Along with acid and alkali based efficient chemical treatment process, physical methods (i.e. shearing, high-pressure homogenization, etc.), biochemical techniques (whole cell-based and enzyme-based) and finally biological techniques (e.g. aerobic and anaerobic treatment) are discussed. During each of the treatment processes, scope of energy recovery and bottlenecks of the processes were elaborated. The review thus provides systemic insight into developing efficient pretreatment processes which could increase carbon recovery and treatment efficiency of PPI waste.

1. Introduction

The major agro-based industries like paper, cotton, sugarcane, vegetable oil, silk, and woolen textiles are identified by the use of a large amount of water consumption and production of large amounts of high energy intensive waste which making highly suitable for coupling through process integration. The agro-based industries mainly revolve around the transformation of agriculture, forestry and fishery derived products. Mostly, the paper industry produces a large amount of emissions that are highly alkaline in nature, thereby altering the pH of both soil and water bodies where they are exuded. Since the complete comprehensive analysis of paper and pulp mill biosludge is not available, it can only be presumed that the lignin concentration in pulp mill biosludge is much higher in comparison to that of municipal biosludge (Table 1). In the following segment, we will be discussing the various types of treatment methodologies that are used in the conversion of this into a useful volume of mass or product substance.

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Table 1
Comparison of parameters for (1) municipal and (2) pulp and paper activated (Xu and Lancaster, 2012).

Parameters	Municipal	Pulp and Paper
Total Solids (TS), %	0.8–1.2	1.0–2.0
Volatile Solids (VS), % of TS	59–68	65–97
Lignin (%)	10–20	5.68
Hemicellulose (%)	20–40	6.53
Cellulose (%)	40–60	32.49
Nitrogen, % of TS	2.4–5.0	3.3–7.7
Phosphorus, % of TS	0.5–0.7	0.5–2.8
Iron, g / kg TS	0	0.33–2.2
pH	6.5–8.0	6.0–7.6
Heating Value, MJ/kg TS	19–23	22–25

2. Pre-treatment strategies

Out of the total paper and pulp biosludge, it was estimated that only about 30–50% of the COD is biodegradable. This is because it contains resistant extracellular polymeric lignocellulosic substances and microbial cells. Multiple pre-treatment methods and techniques have been implemented and studied in order to increase the accessibility to the cellulose polymers and disrupt the floc structure of biosludge which results in an increased anaerobic digestion (AD) and bioresource valorization (Saha et al., 2011; Kaluza et al., 2014). Several methods have been tried and studied under a wide range of treatment techniques, varying from chemical treatments to mechanical/physical treatments.

2.1. Chemical treatment

The two most commonly used alternative conditions for chemical pre-treatment are alkaline and acidic environments. Usually, sludge solubilization is caused and high biogas is produced due to the addition of an alkali to the sludge. But, according to literature, the reports and results are conflicting. After the alkaline treatment, the methane yield reduced by 84% (Bayr et al., 2013). Whereas, on the other hand, it was discovered by Wood et al. (2009), that the yield of methane amplified by 20% and 270% in two distinct types of pulp mill biosludge respectively (Meyer and Edwards, 2014; Saha et al., 2011; Wood et al., 2009). Despite the fact that the increase in soluble CODs generally showed a positive impact on AD, pre-treatment of lignocellulosic materials may be restrained due to some CODs which were reported during this process. For instance, furfural and hydroxymethyl-furfural degradation products from pentoses and hexoses, organic acids and aromatic compounds from lignin in the form of acids, aldehydes and ketones have adverse effects on AD. Under acidic conditions, these compounds are formed even more easily (Bayr et al., 2013; Galbe and Zacchi, 2007). This corresponded to the result established by Bayr et al. (2013) that acid pre-treatment (HNO_3 at pH 3) had an adverse impact on the AD and decreased the methane yield by 99%. Although the results of chemical treatment alone on biosludge appear to have negative or conflicting impacts on AD, there are other studies that have coupled this treatment with other methods. Increasing alkalinity is effective in weakening bacterial cell walls, which allows the other pre-treatment methods to be more effective (Stephenson and Dhaliwal, 2000). Coupled pre-treatments are discussed in the following sections.

2.2. Physical treatment

Mechanical treatment methods are used in order to physically rupture the flocs from within the biosludge so that more of the COD can be made available for biodegradation by the microorganisms. The most commonly used mechanical treatment methods are ball milling, sonication, shearing, and high-pressure homogenization techniques.

NaOH: The effect of pre-treatment can be elevated when NaOH is applied along with the mechanical pre-treatment.

2.2.1. High-pressure homogenization

High-pressure homogenization is the most peculiar and patented mechanical pre-treatment method, in which a pressure of 83 MPa is applied on the sludge and then released quickly through a cell disruption valve. A very high shear force is created onto the sludge by the released pressure, causing disintegration of the sludge flocs into relatively fine particles or soluble COD (Elliott and Mahmood, 2012).

2.2.2. Microwave and ultrasound

Sludge disintegration can be achieved using high-frequency electromagnetic waves such as microwaves and using ultrasound techniques. Small gas bubbles can be formed by using these waves and when these bubbles collapse, strong temperature and pressure gradients are created. This leads to the rupturing of cell walls with intercellular matter being released which can then be more easily degraded by microorganisms during AD (Meyer and Edwards, 2014). Microwave treatment is more effective and efficient than ultrasound treatment. Since microwave treatment is purely a temperature-based treatment, the resulting improvement in AD performance may be caused by heat

Table 2
Summary of mechanical and thermal pre-treatment studies.

Type of treatment	Effects on AD	Reference
High-pressure homogenization (0.1% NaOH, 83 MPa)	34% increase in methane yield	Saha et al., 2011
Mechanical Shearing (1500 rpm)	No significant effect	Elliott and Mahmood, 2012
Sonication (20 kHz, continuous sludge feeding)	15% increase in methane yield	
High-pressure homogenization (0.1% NaOH, 83 MPa)	15% increase in methane yield and significantly faster rate. SRT = 3 days had similar yields as SRT = 20 days in the control	
Wet milling (0.2–0.25 mm balls, 9 min)	20–50% increase in methane yield	Elliott and Mahmood, 2007
Sonication (20 kHz, 30 min)	No significant effect	Wood et al., 2009
Ultrasonic (16.8–39.6 MJ/kg TS, 80 min, 10s on, 5 s off cycles) + Alkaline (0.206–0.261 g/g TS)	0%	Park et al., 2012
Microwave (150 °C)	16%	Tyagi et al., 2014
Ultrasonic (110.2 MJ/kg TS, 60 min)	23%	
Combined alkali (pH = 12) and ultrasonic (110.2 MJ/kg TS, 60 min)	47%	
Ultrasonic (45 kHz, 45 min)	6%	Bayr et al., 2013
Microwave (175 °C)	63%	Saha et al., 2011
Ultrasonic (117.7 MJ/kg TS, 90 min)	51%	
70 °C, 40 min for pulp and paper mill	7%	Bayr et al., 2013
150 °C, 10 min for pulp and paper mill	45%	
170 °C, 1 h for Sulfite pulp mill	54%	Wood et al., 2009
170 °C, 1 h for Kraft pulp mill	467%	

Table 3
Color removal from paper and pulp effluent by white rot fungi.

Name of Strain	References
Phanerochaetechrysosporium and *Trametesversicolor* (40–80% decolorization of kraft mill effluent)	Livernoche et al., 1983; Modi et al., 1998
P. chrysosporium strains and *P. Avidoalba*	Perez et al., 1997
Pleurotusostreatus (69%) and *Pleurotussajor-caju*	Kannan and Oblisami, 1990; Choudhury et al., 1998
Fomeslividus and *T. versicolor* (64–68%) pulp and paper mill effluent	Selvam et al., 2002
Mucoralean fungus *Rhizomucorpusillus* (48–49%) (bleach mill effluent)	Van Driessel and Christov, 2002
P. sajor-caju; *P. platypus* and *P. citrinopileatus* (removal of color from pulp and paper mill effluent)	Raghunathan and Swaminathan, 2004
Klebsiella sp., *Alcaligens sp.*, and *Cronobacter sp.* (SBR for removing pollutant from wastewater ~reduction in color (55%), organic halides (45.4%), TDS (22%) and TSS (86.7%) within 14 h	Kumar et al., 2014

instead of the action of waves or the synergistic effect of microwaves and heat. It has been observed that a lot of energy is required to achieve a significant and major difference in AD performance during ultrasonic treatment (Park et al., 2012) (Table 2). A study was conducted on the energy balance of microwave and ultrasonic treatments. Unfortunately, maximum TS reductions were only 9%, which is not very significant and therefore it is unlikely to make this type of pre-treatment feasible (Saha et al., 2011). Hence, microwave and ultrasonic treatment do not seem to be viable alternative techniques.

2.3. Thermal treatment

The thermal treatment technique is considered an effective pre-treatment method to increase the soluble COD content as well as improve AD (Veluchamy and Kalamdhad, 2017). In the thermal treatment technique, the associated high temperatures and high pressures facilitate cellular disintegration (Elliott and Mahmood, 2007). Norwegian pulp and paper mills used the full-scale thermal pre-treatment technique for the degradation of biosludge (Kepp et al., 2000). With long retention time, sludge solubilization is possible at temperatures as low as 50 °C (Vlyssides, 2004). The high end of the temperature range is approximately 200 °C. Retention time for thermal treatment also varies largely from 0 min to 72 h (Elliott and Mahmood, 2007). Inhibitory compounds may be produced as a result of long thermal treatment. However, it is also reported that thermal treatment for 1 h at about 170 °C does not produce any inhibitory compounds (Wood et al., 2009). There are only two studies that have reported the thermal pre-treatment of pulp and paper mill biosludge and results from studies on other types of sludges are also reported in the summary table below (Table 2). Some studies on thermal treatment mainly focused on COD solubilization and improvement in soluble COD as well as biogas production was achieved (Paul et al., 2006; Vlyssides, 2004).

2.4. Biological pre-treatment strategies

2.4.1. Micro-organisms

The levels of nitrogen and phosphorus are influenced by the microbial decolorization process of pulp and paper mill effluents (Chandra et al., 2011). The routine screening carried out by many researchers of microorganisms capable of treating pulp and paper waste usually depends upon the presence of enzymes that degrade lignocellulosic compounds. Amongst the various microorganisms that were used, the fungi that cause white rot like *Phanerochaetechrysosporium* and *Trametesversicolor* and *P. avidoalba* are well known for decolorizing lignin/phenolic-rich kraft mill effluents as well as helping to reduce absorbable organic halides (AOX) and chemical oxygen demand (COD) of discharged materials (Livernoche et al., 1983; Modi et al., 1998). These white rot fungi are well-known producers of extracellular isoenzymes, lignin peroxidase, MnP-dependent peroxidases (MnPs) and laccases, which are capable of degrading lignins as well as the chlorinated lignins found in pulp bleaching effluents (Kirk et al., 1986; Lankinen et al., 1991). Purified enzymes such as Lignin peroxidase and Manganese peroxidase were found to be more efficient and effective in the decolorisation of kraft bleach plant effluent and olive mill wastewaters (Sayadi and Ellouz, 1995; Perez et al., 1997). Fungal strains e.g.*Trametesversicolor* (ATCC 20869); *Coriolusversicolor*; *Pycnoporuscinnabarinus*; *Trametesvillosa*; *Aspergillus fumigatus* VkJ2.4.5; *Aspergillus fumigates*; *Pycnoporussanguineus* and bacterial strains eg. *Streptomycescyaneus* CECT 3335; *γ-proteobacterium* JB have been reported as laccase producersused for biobleaching of pulp (Singh et al., 2015). Laccase obtained from various microbial species caused significant changes in the color remediation and the toxicity of these samples. Laccase from *T. versicolor* alone caused 82% COD reduction with 99% of 2,3,4,6-tetrachlorophenol (2,3,4,6-TCP), 98% of 3,4-dichlorophenol (3,4-DCP), and 77% of 4-chlorophenol (4-CP) removal from paper mill effluent (Pedroza et al., 2007). The deep brown effluents from paper and pulp mills can be decolourized by pure laccase from *T. versicolor* (Karimi et al., 2010). Similarly, laccase obtained from *T. versicolor* through solid-state fermentation of brewer's spent grain demonstrated significant decolourization (~87.7%) of various structurally different industrial dyes (Dhillon et al., 2012). *Pleurotusostreatus* and *P. sajor-caju* have been established for color removal of mill sludge (Kannan et al., 1990; Choudhury et al., 1998). The other two microorganisms that were used for the treatment of pulp and paper mill effluent are *Fomeslividus* and *T. versicolor* (Selvam et al., 2002). In the decolorization of bleach mill effluent, Mucoralean fungus *Rhizomucorpusillus*was also reported (Driessel and Christov, 2002). Raghunathan and Swaminathan, 2004, in 2004 observed that *Pleurotus* spp. eg. *P. Sajor-caju*; *P. platypus* and *P. citrinopileatus* release inorganic chloride (524–814 mg/dL) during removal of color from pulp and paper mill effluent. They observed that the efficiency of color removal (~60%) and COD removal (57.2%) was better in *P. sajor-caju* than in *P. platypus* and *P. citrinopileatus*. For the treatment of wood-based pulp and paper industry wastewater, algae can also be used by removing up to 58% of COD, 84% of color and 80% of AOX (Tarlan et al., 2002) (Table 3).

2.5. Enzymatic pre-treatments

The current requirement for customer satisfaction has increased the demand for chlorine-free bleached pulp (Garcia et al., 2010; Sharma et al., 2014). But, with the help of potent biocatalysts such as xylanases and laccases, eco-friendly biobleaching, pitch removal and de-inking of paper waste is possible. When this process was the first used it was not considered technically and economically feasible due to the lack of availability of these biocatalysts. Other limitations were that these biocatalysts were too costly and cumbersome. Enzymes like laccases are required as mediators for their bleaching action (Singh et al., 2008, 2011a, 2011b). But, recently companies that producer enzymes have developed and improved biocatalysts to offer significant benefits to the pulp and paper industry (Bajpai, 1999). Chemical techniques like (1) bleaching of pulps with oxygen and extended cooking, (2) hydrogen peroxide, and (3) ozone treatment maybe options. But most of these methods are too expensive. Thus, the use of enzymes will be an alternative and more cost-effective technique to minimize the utilization of chlorine and other similar bleaching chemicals (Bhoria et al., 2012).

2.5.1. Role of hemicellulases (xylanase)

In the past decade, particularly because of their biotechnological potential in various industrial processes, such as food, feed, and pulp and paper industries, xylanolytic enzymes have attracted a great deal of attention (Beg et al., 2001). Xylans are intimately linked to cellulose and lignin and are present within the pulp. Endoxylanases and β-xylosidases (collectively called xylanases) are the two key enzymes responsible for hydrolysis of xylan, the chief component of hemicellulose (Chávez et al., 2006). Endoxylanases act on the homopolymeric backbone of 1,4-linked β-d-xylopyranose producing xylooligomers (Ahmed et al., 2009) while β-xylosidases act on these xylooligomers releasing xylose (Knob et al., 2010). In nature, hemicellulases are produced by a wide variety of microorganisms, including bacteria and fungi (Ahmed et al., 2009). Worldwide, the most commonly used enzyme for prebleaching technology is xylanase. When xylanase hydrolyzesxylan polymer, during the bleaching action the xylan backbone affects their separation. Xylanase helps to increase fibre wall swelling and in turn increases the speed of diffusion through the walls (Clark et al., 1991). Extraction is facilitated by covalent binding of lignin to xylan to start enzymatic degradation. Xylanase enzymes also catalyze the hydrolysis of xylan that has re-precipitated on the fibres during alkaline pulping. The efficiency of xylanase treatment can be influenced by various factors such as pH (pH 5–11), temperature (35–60 °C), enzyme dosage, pulp consistency, and reaction time. Xylanase was found to be completely stable over a broad pH (5–11) range and retained 52% of its activity upon incubation at 70 °C for 30 min (Nagar et al., 2011). Xylanases have been widely tried in the pulp and paper industry since they showed lignin extractability from kraft pulp by de-polymerizing xylan closely associated with lignin in the plant cell wall. Examples of commercial xylanases and processes are Novozymes 473, Cartazyme HS-10, reduced chlorine consumption by 31% and increased the final brightness by 2.1–4.9 points (Bajpai et al., 1994). Xylanase P (10Ug − 1 pulp) improved the brightness of kraft pulp by 5.6 points and caused a 10% decrease in chlorine consumption (Madlala et al., 2001). Xylanase from *Bacillus megaterium* showed 8.12% and 1.16% enhancement in brightness and viscosity and 31% reduction in chlorine consumption (Sindhu et al., 2006). Xylanase from *B. stearothermophilus* SDX reduced chlorine consumption by up to 15% while its combination with pectinase resulted in a 20% reduction (Dhiman et al., 2008). Alkali stable and thermos-tolerant xylanase from *Bacillus pumilus* SV-85S showed elevated and better brightness by 1.9 points. The pre-treatment of pulp with xylanase resulted in a 29.16% reduction in chlorine consumption while maintaining the same brightness as in the control (Nagar et al., 2013).

2.5.2. Role of laccase

Laccase is a versatile biocatalyst that plays a significant role in the delignification and brightening of pulp as well as removing lipophilic extractives responsible for pitch deposition from both wood and nonwood pulps. Therefore, by either forming reactive radicals with lignin or by functionalizing lignocellulosic fibres, laccases are capable of improving physical, chemical as well as mechanical properties of pulp. Laccases can also target the colored and toxic compounds released as effluents from pulp industries during polymerization and depolymerization reactions (Virk et al., 2012). Laccases have an advantage over other lignin-oxidizing enzymes, like lignin peroxidases (Lip) in that the redox potential of Lip increases with a decrease in pH of the reaction environment (Singh et al., 2009; Canas and Camarero, 2010). Since several steps of the papermaking process pass through alkaline conditions, the pulp and paper industry requires alkali tolerant laccases for delignification purposes (Singh et al., 2008, 2011b). The delignification mechanisms of xylanases and laccases are dissimilar because xylanases help to improve the delignification by making pulp more vulnerable to attack by bleaching chemicals whereas laccases act directly on lignin and cause its extraction from pulps (Virk et al., 2012).

2.5.3. Role of enzymes in pitch control

Pitch is composed of a variety of fatty acids, sterols, resin acids, glycerol esters of fatty acids and other fats and is usually considered as the wood component that is soluble in methylene (Allen, 1975). Particular types of wood pulps such as sulfite pulps and various mechanical pulps, especially from pines are known to have high pitch contents (Gutiérrez et al., 2001). Lipidase 10,000 (American Lab. Inc.) and Candida and Aspergillus lipases have been investigated and researched for the enzymatic control of pitch (Virk et al., 2012).

2.5.4. Role of cellulolytic enzymes

Cellulolytic enzymes are one of the most important industrial enzymes that have demonstrated their biotechnological potential in various industrial sectors like food, brewing, wine making, agriculture, pulp and paper (Pandey et al., 1999; Kuhad et al., 2011). Cellulase is not a single enzyme but is a complex of three major and different types of cellulases: endoglucanases, exoglucanases, and cellobiase. Bioconversion of lignocellulosic waste to soluble sugars relies on the synergistic action of the complete cellulase system comprising of CBH, EG, BG, and hemicellulases (Aro et al., 2005).

3. Biological treatment strategies

Nowadays, biological treatment systems are commonly used as an alternative option for the reduction of BOD and COD as well as the color of effluent as physical and chemical processes are not capable of removing biological oxygen demand (BOD) and low molecular weight compounds (Singh and Singh, 2004). Across the world, there are multiple biological treatment processes that exist for paper and board mill effluent such as those based on aerobic, anaerobic, algal and fungal biomass treatment. Aerobic treatments are aerated lagoons, activated sludge systems and biofilm processes that need oxygen (Persson, 2011).

3.1. Aeratedlagoon

One of the most commonly used and simple types of biological treatment is aerated lagoons, which is are large, shallow (≈ 4 m) ponds where wastewater is treated with active microorganisms and mechanical aeration. But, this consumes lots of energy for aeration and space for a residence time of 5–7 days. However, due to its easy operation and capacity to withstand higher shock load, the preferred option is conversion into LAS-facilities that function as long-term aerated activated sludge treatment plants with a lower residence time (1 day). Parts of the old aerated lagoon can then been utilized for aeration and other parts can be used for temperature stabilization and cooling of hot wastewater effluents. The aerated lagoon will primarily eliminate BOD but can also reduce emissions of AOX to some extent. The removal efficiency is dependent on residence time, pH, temperature, amount of sludge and degree of aeration (Persson, 2011).

3.2. Activated sludge process

The pulp and paper industry is a major producer of wastewater. In a pulp and paper mill, the primary sludge is mainly composed of wood fibres and chips, sometimes called fibre sludge. The wastewater then flows on to an aerobic treatment process(also called activated sludge process) where aerobic microorganisms digest components and particles of the wastewater and produce large amounts of biomass in the process(referred to as waste activated sludge (WAS) or biosludge) (Ashrafi et al., 2015). Though suspended solids can be removed by primary treatments, secondary treatment is required in order to remove dissolved organic compounds and colloidal particles from wastewater

(Thompson et al., 2001). During secondary biological treatment, dissolved organic compounds and colloidal particles are consumed by microorganisms and oxidized into low molecular weight fractions. Activated and advanced wastewater treatment strategies are necessary to degrade lingo-cellulosic compounds and halogenated chemicals in effluents that pose the greatest threat to human health. During the activated sludge process, wastewater is treated in two steps: aeration and sedimentation. In the first step, nonspecific microorganisms were used to treat wastes coming from the pulp and paper industry using alow food/microbe (F/M) ratio, powerful tank aeration and retention times that vary from a couple of hours up to a day. This technique helps to regulate the treatment plant using high mixed liquor suspended solids (MLSS) to tolerate sudden shock, buffering the BOD, stabilizing pH, controlling wastewater composition and regulating temperature. In the second step, water and sludge are separated in a sedimentation basin and parts of the sludge are pumped back to the aeration basin. The activated sludge process used in wastewater treatment, produces a large quantity of waste activated sludge (WAS) or biosludge, which has a dry solid composition of approximately 0.5–2%. The recirculation of sludge with a greater load of microorganism has importance for the extensive reduction in organic material. Compared to the aerated lagoons, these activated sludge systems are more sensitive and cannot stand fast load changes. However, the degree of efficiency can be controlled and very high BOD-reductions were observed and noted (Thompson et al., 2001; Persson, 2011). The microorganisms: bacteria, protozoa, fungi, and rotifers exist in the activated sludge reaction vessel and clarifier as floc.

A low F/M ratio leads to inadequate food for the population of microorganisms causing problems in maintaining sufficient dissolved oxygen (DO) concentration. This ultimately leads to improved growth of filamentous microorganisms. However, such systems are usually less effective in removing color, COD and chlorinated phenolic compounds (Raj et al., 2007). Therefore, more advanced, flexible and effective alternative biological wastewater treatment strategies like SBR have been used by many researchers even though high concentrations of toxic compounds were formed (Mahvi et al., 2004 and Tsang et al., 2007). The effect of MLSS concentration and high volumetric organic loading (6.67 kg COD/m^3 d) was studied with an SBR system under the most favorable condition of 5 h per cycle aeration time with HRT 24 h and average TDS and TSS removal efficiencies above 71% (Kumar and Subramanian, 2014). Pulp and paper industrial effluents are a rich carbon source that are deficient in N and P (Dijkstra et al., 2011). For effective treatment of wastewater, a significant quantity of N and P must be added because microorganisms present in the effluent require N and P to produce enzymes for the degradation of organic matter. If quantity of N present is not sufficient, it can result in the filamentous and dispersed growth of a microbial population which settles poorly. As a general rule, the ratio of N and P required with respect to BOD load is 100 BOD: 5 N: 1 P.

3.3. Moving bed bioreactor

The Moving Bed Bioreactor (MBBR) is made of thousands of suspended plastic carriers that float free with microorganisms attached as films on the carriers. Air is supplied from the bottom of the reactor which keeps the carriers moving and permits a higher load, better mixing, and higher flush. The biggest advantages of a suspended biofilm process is that it does not need the return of activated sludge, has very small space requirements, has high-quality shock-resistance and can operate at very high concentrations of biomass (Persson, 2011).

3.4. Biofiltration

The biofilter reactor is a tertiary treatment process, which is constructed with a biological filter of a fixed biomass carrier and a biological contactor. Wastewater and air are supplied from the bottom of the reactor and led in an upward direction through the dense granular bed. The filter is operated with a hydraulic retention time of around 0.5 h, and very high BOD reductions are observed. But, due to clogging problems at higher BOD concentrations, wastewater containing low BOD concentrations can only be used as a substrate (Möbius, 2006).

3.5. Anaerobic treatment

Anaerobic treatment is more appropriate for treatment of high strength paper and pulp industry wastewaters but is not used as widely as the aerobic treatments (Ochre-Media, 2001). Effluents originated from recycled fibres are often treated anaerobically. Compared to aerobic treatment, this technique has many advantages: lower sludge production, lower chemical consumption, smaller space requirements and energy production in the form of biogas (Persson, 2011). The major problem of anaerobic treatment for pulp and paper effluents is hydrogen sulphide formation since sulphate is widely used as an active cooking chemical in many pulp mills. The anaerobic process is sensitive to toxic compounds present in wastewater (Thompson et al., 2001). This kind of system provides a source of energy with no net increase in atmospheric carbon that contributes to climate change. Energy produced through the anaerobic digestion process can help reduce the demand for fossil fuels. It is particularly suitable for treating low pollution loadings (SS $<$ 200 mg/L and soluble chemical oxygen demand (COD) $<$ 400 mg/L). Since pulp and paper mill biosludge is more difficult to digest than municipal biosludge, many studies have focused on using different methods to enhance the AD process instead of simply digesting the raw biosludge. Co-digestion and different pre-treatments have been discussed by many scientists to improve the biodegradation of pulp and paper mill sludge (Table 4). There are a wide range of substrates used in co-digestion experiments, such as municipal sludge, food waste, dairy farm waste, rice straw, pig slaughterhouse waste, grease trap waste, grass silage, and pulp mill sludge (Borowski and Weatherley, 2013; Karthikeyan et al., 2016; Chakraborty et al., 2018; Chakraborty and Venkata Mohan, 2018; Hagelqvist, 2013; Mussoline et al., 2013; Trulli and Torretta, 2015; Yalcinkaya and Malina Jr., 2015). It was observed that by the addition of a small amount of food waste a significant increment in the biogas generation was observed (Yun et al., 2015). This is consistent with the idea that a limiting factor in pulp and paper mill biosludge digestion is limited nutrients for microorganisms that food waste is able to supplement. In the study by Huilinir et al. (2014), natural zeolite was used as a catalyst and although this is not co-digestion of two substrates, it is consistent with the addition of dissimilar material to the biosludge to enhance its AD.

Table 4
Summary of co-digestion of pulp and paper mill biosludge with other substrates.

Co-substrate with pulp and paper mill biosludge	Amount added	Operating conditions	Enhancement over biosludge alone (specific CH_4 yield)	Reference
Food Waste	10% of total COD	Batch	55% increase	Yun et al., 2015
Municipal Biosludge	Up to 50% of total VS	Batch	~50% increase	Hagelqvist, 2013
Monosodium glutamate waste liquid	25% of total dry mass	Semi-continuous	245 mL CH_4/gVS	Lin et al., 2011
Natural Zeolite	0.2–20 g/L	Batch	Maximum 10% increase	Huilinir et al., 2014

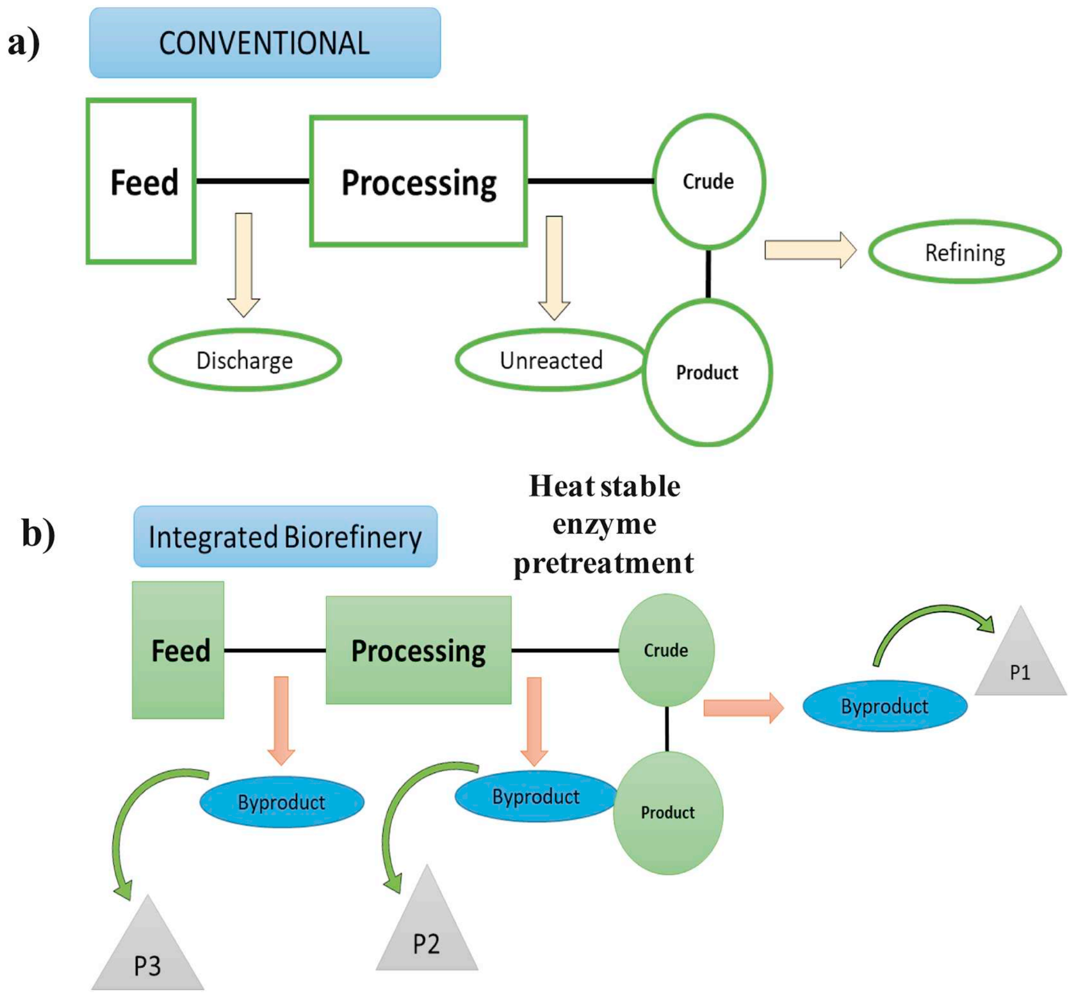

Fig. 1. Comparative a) conventional b) integrated lignocelluloses degrading heat stable enzyme cocktail pretreatment strategies of paper and pulp lignocellulosic biosludge. Where, P1, P2 and P3 referred to the targetted products/bioresource can be recovered during integrated recovery.

4. Bottlenecks of conventional methods

The complete potential of PPI waste utilization will come into play, only when all energy-rich components (including lignin) are fully researched and utilized. However, the literature suggests many limitations in pretreatment followed by energy recovery.

4.1. Drawbacks of incineration and land filling

Mechanical dewatering processes are not able to increase solid content beyond (20–35%) unless more costly dewatering methods are used (Kyllönen et al., 1988). The main drawback of incinerating the dewatered sludge is a low net energy yield of 2–6 MJ/kg (Stoica et al., 2009), which is lower than the burning of wood which yields roughly 17–21 MJ/kg (Smil, 2008). The dewatered sludge cannot be used as fertilizer because it contains heavy metals and other contaminants that are above the regulatory limits. Secondly, there is a generally negative perception of sludge from farmers contributed by the odor of sludge and its impact on neighbors (Meyer and Edwards, 2014).

4.2. Inhibition of AD process and high HRT

It is believed that AD of PPI biosludge is hindered by nutrient deficiency and lignocellulosic and sulfur-containing substances (Hagelqvist, 2013). The hydraulic retention time (HRT) for AD usually has long residence times > 20 days, which result from the slow hydrolysis phase (Kaluza et al., 2014). Longer retention times, as well as the high water content in the sludge, means larger digesters and higher capital cost. This is another reason that AD of pulp and paper mill sludge is still in its infancy (Meyer and Edwards, 2014).

4.3. Low biogas yield

Previous studies have shown that the AD of PPI biosludge has low biogas yield compared to that of other sludges. The methane yield of biosludge in a municipal wastewater treatment plant is between 325 and 380 mL CH_4/g VSadded, whereas the reported methane yield of pulp and paper mill biosludge peaks at 199 mL CH_4/g VSadded and can be as low as 50 mL CH_4/g VSadded (Huilinir et al., 2014).

4.4. Recalcitrant compound rich PPI sludge

Conventional pre-treatment of recalcitrant and/or toxic compounds of PPI sludge showed no improvement of AD performance and in some cases had a detrimental effect even though the soluble COD content had improved. Characterization of those recalcitrant and toxic compounds is very important for a better understanding of the effects of pretreatments and there are no reports on the molecular basis of characterization till date. Of course, this is an extremely difficult task due to the complexity of biosludge.

4.5. VFA toxicity and inhibition of methanogenesis

The soluble COD released after the pretreatments of PPI increases the VFA concentration which also increases the rate of methanogenesis (Xu et al., 2014; Chakraborty et al., 2018; Chakraborty and Venkata Mohon, 2018). However, a drop in pH can cause the reactor to fail if there is not enough buffering capacity to maintain a fairly neutral pH (6.8–7.2) to allow the methanogens to function normally (Kerri, 1999). To increase biogas production and solids reduction, the efficiency of pre-treatment methods need to be improved by using enzymes and ozonation which have a cost associated with them. Therefore, even though treatment may have positive impacts on AD, it may not be economically feasible in full-scale operation. Thus, advanced integration of economic pretreatment methods is required in order to make a controlled release of wide stream of sugars (glucose, galactose, mannose, xylose and arabinose) which can make any of these methods economically feasible and increase the probability of enhancement in bioresource recovery.

4.6. Prospect of the thermostable enzyme pretreatment of PPI waste

Enzymatic pretreatment has important advantages as it consumes low chemical and energy inputs, with no need for corrosion resistant processing equipment, less acid waste and the potential as a promising option for almost complete conversion to replace the well-established thermochemical or physicochemical conversion technologies. Each of these methods is targeted towards uses of enzymes e.g. peptidase, carbohydrolase and lipase, in addition to heat and other chemicals, to systematically convert the carbohydrate portion of the PPI waste (hemicellulose and cellulose) into an intermediate sugar stream that can then be fermented into a range of advanced biofuels and value-added chemicals. Significant research is being performed to optimize bioconversion processes targeting different types of wild-type or engineered microorganisms. But, a new generation of enzymes and enzyme production technologies are needed to cost-effectively hydrolyze the PPI waste. Thus, there is a promising future for the use of heat-stable enzymatic pretreatment for bioconversion processes. But this can only be a reality if the focus is on producing cheap and naturally occurring heat-stable enzymes and then use of a variety of techniques to increase their efficiency to decrease the cost of the enzyme unit operation in the sugar extraction process.

5. Conclusion

Global paper consumption and PPI industrial waste production is increasing day by day and may go up to 500 million tons/year by 2025. Most of the fraction of PPI waste not treated properly using the conventional treatment processes is due to the presence of lignin-rich compounds that get wasted. Nontoxic lignin conversion could be an attractive industrial option for bioresource recovery. Conventionally, two of the most used treatment processes are landfill and incineration and both these options entail costs for the sector. To minimize the cost and maximize conversion into profits, the lignocellulosic part of PPI waste needs to be converted for complete recovery of the organics of PPI waste. However, lignin degradation is a rate-limiting step and may be successfully converted by using the efficient integration of physico-biological treatments. To overcome this challenge, the integration of efficient lignocellulosic conversion technology with a biorefinery approach could be a worthwhile step. Heat stable enzymes need to be considered to overcome the limits of accessibility of microbial enzymes to other major cell wall polysaccharides. The schematic summarized in Fig. 1 helps to visualize how the PPI intermediate products can be utilized to their full potential. Utilization of lignocellulosic byproducts could improve the sustainability of the bioenergy chain and reduce the negative environmental impacts related to inappropriate disposal.

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