Review Article

Different Type of Paper Pulping and Process of Green Bleaching

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Abstract

Biocatalysis is currently considered as a useful alternative to conventional chemical process technology in industry. This is because unlike chemical catalysts, enzymes (the biological catalysts) have the advantage of accomplishing complex chemical conversions under mild environmental conditions of pH, temperature and pressure with high specificity and efficiency. The enzymatic processes are in general eco-friendly (also referred to as green technologies). High degree of substrate specificity eliminates the production of undesirable side products. The bulk enzyme utilization in different industries reveals that over 45% of the enzymes produced are being used in the food industry and the remaining is shared by detergent (34%), textile (11%), leather (2.8%), paper and pulp (1.2%) and other industries (5.6%) excluding enzymes in diagnostics and therapeutics. Paper and pulp industry in India is one of the major contributors to environmental pollution. The conversion of wood into paper involves two processes: Pulping & Bleaching, both of which involve delignification.

Keywords: Xylanase, Xylan, Biobleaching, Eco-friendly, Paper pulp

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Introduction

Lignocelluloses are the most abundant inexhaustible natural materials in the biosphere accounting for more than 60% of total biomass produced [1]. lignocellulosics contain cellulose (insoluble fibres of β-1, 4-glucan), hemicellulose (non-cellulosic polysaccharides including xylan, mannans, galactan and arabinan) and lignin (a complex polyphenolic structure) interacting with each other via covalent and non-covalent linkages, with xylan being found at the interface between lignin and cellulose [2, 3]. The chemical properties of the components of lignocellulosics make them a substrate of enormous biotechnological value. Large amounts of lignocellulosic waste are generated through forestry and agricultural practices, paper-pulp industries, timber industries and many agro industries and they pose an environmental pollution problem. Over 75% of organic chemicals are produced from five primary base-chemicals: ethylene, propylene, benzene, toluene and xylene which are used to synthesize other organic compounds, which in turn are used to produce various chemical products including polymers and resins [3].

Pulping and their types

The pulping process primarily aims at removal of lignin & to separate wood fibers from each other. When the pulp is used to manufacture white papers, it is necessary to continue the delignification until the pulp has less than 1% lignin and If the pulp is to be used for the manufacture of container board, its lignin content may be reduced to 8-10%, usually pulping process is stopped at a lignin content of 4-5% because further treatment under these conditions results in unacceptable losses in cellulose or degradation of the mechanical properties of the fibers. Pulping is the first step of paper manufacture process in which separation of cellulose fibers from the lignin components [2]. In paper making process the break and subsequent removal of all lignin present: a) pulping processes during which lignin is removed by the heating process; b) the residual lignin remains blocked on the cellulose fiber surface (mainly linked to short-chains xylan), is then removed by a multistep bleaching process by the use of large amounts of chlorine (Cl_2) and chlorine dioxide (ClO_2) chemicals. The difficulty to remove lignin from the cellulose fibers in that xylan forms a lignin-carbohydrate complex with lignin, acting as a physical barrier that prevents the bleaching agent's action on the lignin. The pulping process may involve either chemical pulping or chemi-mechanical pulping as depicted below:

Chemical Pulping

This process utilizes large amounts of chemicals in the presence of heat and pressure to remove the lignin that binds the cellulose fibers together. The pulp produced by this process is used in the manufacturing of high-quality paper which requires high brightness or high mechanical strength. The purpose of any chemical pulping process is to facilitate the separation of the individual fibers by the dissolution of lignin [4]. It may be of three type viz. Kraft pulping, sulfite pulping & soda pulping. Most of the integrated pulp and paper mills in India and abroad adopt the cost-effective Kraft process for pulping.

Kraft Pulping

The Kraft pulping process introduced in 1879 and it was invented in Germany by Carl F. Dhal [1], is still the dominant method [4]. The Kraft word in German language means strength implicating that paper made by Kraft pulping process is superior in strength when compared to other processes. Around 70% of world's pulp is still made by this process [1]. Kraft pulping involves three main steps: digestion (wood chips are cooked), washing (black liquor is separated from the pulp) and chemical recovery (chemicals are recovered from the black liquor for reuse). In the digestion process, wood chips are preheated in a steaming vessel to remove air and assure good liquor permeation and digested into an anaerobic alkaline environment (with a mix of NaOH & sodium sulphide at 165˚C for 60 min) resulting in solubilization and removal of lignin, while minimizing degradation of cellulose or loss of mechanical properties of the fibers. In this process, the short chain xylan precipitates with its re-adsorption on the top of cellulose microfibrils. The kraft process stops when the lignin content remains 4-5 % as further treatment under these conditions results in unacceptable losses in cellulose or mechanical properties of the fibers. The remaining ~5% lignin imparts brown color to the pulp. The yield of pulp is about 45%. The process has a recovery system that not only recycles the pulping chemicals but is also a source of about one-half of the steam generation. Kraft process has many advantages over other pulping methods i.e. a wide range of hardwood & softwood can be pulped, cooking time is shorter, less pitch is generated, the pulp has good strength properties, and spent liquor is more easily recovered.

Sulfite pulping

In this process, wood chips are cooked under acidic conditions with a mixture of sulfurous acid and bisulfite. It produces pulp of lower physical strength and bulk, but exhibits better sheet formation properties. The yield on the basis of chipped wood is again about 45%. These pulps are blended with ground wood for newsprint and are used in printing, bond papers, and tissue.

Soda pulping

Soda pulping is an alkaline process involving two phases that are the solid and the liquid phase. Soda pulping is the first pulping method adopted industrially as per the historical evidences. This process has some drawbacks such as longer cooking time, low pulp yield, high temperature and low paper strength. The soda pulping is used for the conversion of agro residues (like wheat and rice straw and bagasse) to pulp. It involves treatment with soda and water at 174˚ C for 45 minutes. Soda pulping method is a principal practice method as far as cooking of agro residues is concerned [5]. Use of anthraquinone (AQ) enhances the outcomes of soda pulping process as it accelerates the delignification rate [6, 7]. Soda-AQ process even at low cooking temperature increases the pulp yield and provides pulp with good strength properties. Soda-AQ pulping is the most preferable method for the conversion of non-wood raw materials to fibrous pulp [8]. In contrast to soda process, desired kappa number can be achieved at low cooking time and low temperature conditions in soda AQ process.

Mechanical pulping

It uses mechanical force and pulp retains much of the lignin. This process requires a lot of energy and results in damage to the fibers. Thus, two important goals are sought in advancing this technology. The first is to reduce the energy consumption in the mechanical refining. The second is to reduce the damage to the fibers. Recent developments have resulted in advances in both areas. The yield of pulp is over 90%. The pulp prepared from this process is used in manufacturing papers that do not require high strength. In refining pulping steam pre-treatment is done that gives thermo mechanical pulp. This mechanical pulp is yellow in colour due to high quantity of lignin [9].

Biopulping

Biopulping is the treatment of wood chips and other lignocellulosic materials with natural wood decay fungi prior to thermo-mechanical pulping. When wood chips are pre-treated with an inoculum of fungi prior to relining, the wood chips become softened and more porous. Wood is debarked, chipped and screened according to normal mill operations. Then chips are briefly steamed to reduce natural chip microorganisms, cooled with forced air, and inoculated with the biopulping fungus. The inoculated chips are piled and ventilated with filtered and humidified air for 1 to 4 weeks prior to processing. The main advantages of biopulping are reduced electrical energy consumption (at least 30%), improved paper strength properties and reduced environmental impact. The biopulping is technologically feasible and cost-effective [10].

Bleaching of Pulp

Bleaching refers to the purification and whitening of pulp. The purpose of the bleaching process is to enhance the physical and optical qualities (whiteness and brightness) of the pulp by removing or decolorizing the residual lignin (~ 5%) and oxidizing residual chromophores so that they can be solubilized during extraction to give a white pulp of high brightness. This process creates an oxidizing environment that can both delignify and bleach. Bleaching sequence is a combination of a series of stages, each stage involves three steps i.e. mixing of pulp with chemicals and heat, retention of the mixture in suitable vessels and washing of the pulp with water. Different stages in bleaching are as follows:

Chlorination

Chlorination is the treatment of pulp with chlorine gas or chlorine water. This process converts lignin to compounds (by oxidation and substitution) that are soluble in water and alkali. It removes bulk of the liquid. This step is often performed in conjunction with alkali extraction.

Alkali (NaOH) Extraction

This step dissolves the degraded lignin. The alkali extraction facilitates neutralization of acidic groups and removes chlorinated resins & hemicelluloses.

Chlorine Dioxide Treatment

Chlorine dioxide is often used for bleaching either at the chlorination stage (as a substitute for some of the chlorine) or as an additional chlorine dioxide stage. Chlorine dioxide has greater oxidizing power than chlorine and is used for nearly all high brightness pulps. Chlorine dioxide oxidatively degrades the remaining lignin and destroys other chromophores in the pulp.

Effect of Chemical Bleaching on Environment

Chlorinated compounds generated during bleaching process are released into the environment with the wastewater from the mills leading to pollution. The presence of these compounds in the effluents increases their COD values ultimately resulting in toxicity to aquatic life as well as enhances the cost of effluent treatment. The by-products formed during chemical bleaching are toxic, mutagenic, carcinogenic, persistent, bioaccumulating and cause numerous harmful disturbances in biological systems. In response to government and environmental protection groups, paper industries are currently changing practices to reduce the expulsion of effluent loaded with BOD, COD, total suspended solids and colour through new wastewater treatment techniques [11]. The pulp and paper industries are setting its goals on in-situ process modifications by addition of oxygen delignification, extended cooking, replacement of chlorine with chlorine dioxide (ClO_2) and use of strong oxidizing agents like hydrogen peroxide and ozone [12].

Biobleaching

Biobleaching which involves the use of microbial enzymes such as xylanase & laccase, is an effective alternative to reduce the environmental pollution. At present, xylanases are being used by majority of paper and pulp industries, particularly abroad. In India, it is still in infancy stage due to non-availability of cost-effective xylanase having desirable properties. Laccase (E.C. 1.10.3.2.), an N-glycosylated extracellular blue copper oxidase, is produced by

most of the white-rot fungi. Laccase is being used primarily for the degradation of lignin in paper and pulp industry. Biobleaching of pulps using xylanases is one suitable alternative to be used in the pulp and paper industry to reduce or eliminate the use of chlorine and chlorine dioxide, as well the operational costs related to water, electricity, and fuel [13]. Major advantages of biobleaching are: reduced consumption of bleaching chemical, reduced absorbable organic halogen compounds, improved pulp and paper quality, improved brightness, reduced effluent toxicity and pollution load [11].

Xylanase as a Bio bleaching Agent

Xylanase (endo-1,4-β-D-xylanohydrolase; E.C. 3.2.1.8) depolymerises xylan by random hydrolysis of the xylan backbone to produce shorter xylo-oligosachharides (containing 2 to 5 xylosyl residues), xylobiose and xylose. β-xylosidase hydrolyses xylo-oligosachharides and xylobiose from their non-reducing end to produce xylose. Xylan is the second most abundant biopolymer after cellulose & main hemicellulosic component. It accounts for 20-25% and 7-12% of total dry weight of hardwoods and softwoods, respectively.

Xylanase is an industrially important enzyme having applications in paper and pulp industry, fruit juice and wine clarification, food industries, extraction of plant oils, texture improvement of bakery products, production of xylose, textile industry, bioconversion of agricultural wastes, retting of plant fibers and improvement of nutritional value as poultry feed [14]. Viikari *et al.* [15] were the first to demonstrate that xylanases could be useful in pulp and paper industry. In paper and pulp industry, xylanase finds immense application as a biobleaching agent. The potential biotechnological applications of xylanase in pulp and paper industry have been reported by several workers [16, 17].

In the pulp and paper industries, the use of cellulase-free xylanase for bleaching purposes is better for the environment, as chemical bleaching uses chlorine-based chemicals that are toxic [18]. Xylanases used for this process must be highly active at alkaline pH [19]. The production of cellulase-free xylanase is very important for their application in the paper industry [20] as the presence of cellulase may destroy cellulose fibers present in the pulp.

Process of Biobleaching with xylanase

It involves enzymatic pre-bleaching of pulp followed by chemical bleaching (**Figure 1**). In enzymatic pre-bleaching, extensively washed pulp of 10% consistency is pre-treated with an optimum concentration of xylanase (crude or purified) in a plastic bag and under optimum conditions of temperature and time in a water bath. After enzyme treatment, the pulp is de-watered followed by washing with cold water and dried at 80°C for 24 h. A control sample (not pre-treated with xylanase) is also processed under identical conditions. The enzyme dose, temperature and time used for pre-bleaching are optimized for best results. Then, the pulp is subjected to chemical bleaching sequence ($CDED_1D_2$) under the conditions specified in **Table 1**. The dose of chlorine treatment given to xylanase-pre-treated pulp is reduced progressively to the level so as to achieve the same brightness as in control, keeping all other parameters constant. After bleaching treatments, pulp pads are made which are analysed for brightness, viscosity and paper strength parameters.

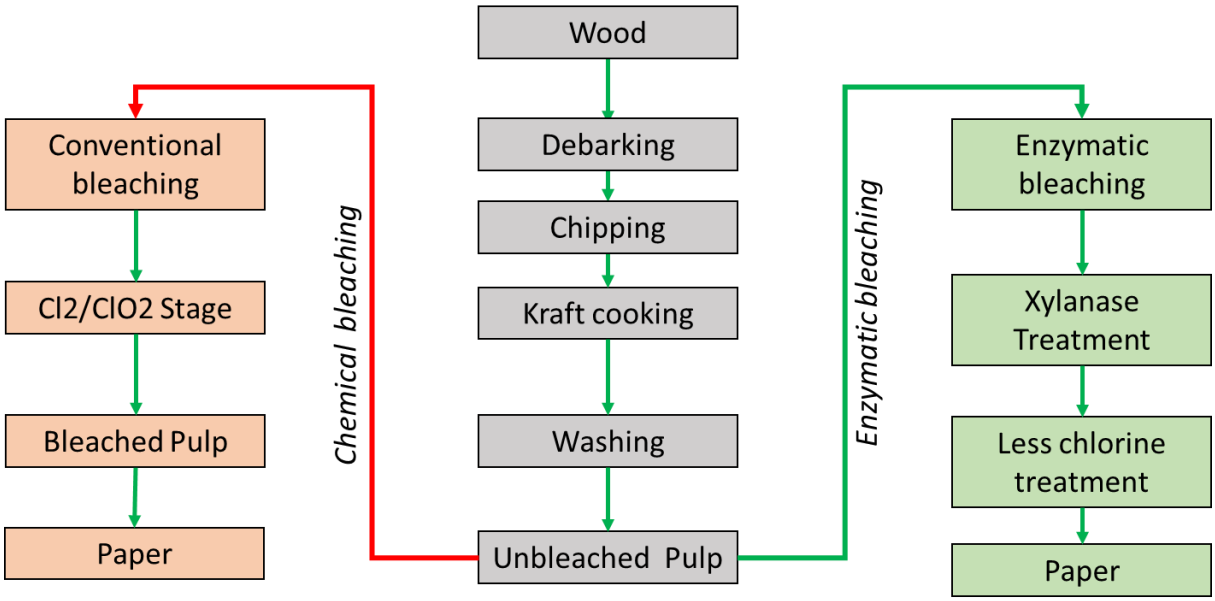

Figure 1 Flow chart showing kraft pulping via chemical and enzymatic bleaching process

Table 1 Conditions for chemical bleaching sequence ($ECDED_1D_2$) of mixed wood pulp

Treatment	Pulp consistency	Temperature (˚C)	Time (min)	pH
E (enzymatic prebleaching)	10%	55	120	7.0
CD (chlorine-chlorine dioxide bleaching)	3%	Ambient	45	1-2
E (alkali extraction)	10%	65-70	120	11-12
D_1 (chlorine dioxide stage 1)	10%	70-75	180	3.8-4.0
D_2 (Chlorine dioxide stage 2)	10%	70-75	180	3.8-4.0

Role of Xylanase in Biobleaching

In biobleaching, xylanase is being used, primarily, for the removal of lignin-carbohydrate complexes (LCC) that are generated in the kraft process and act as physical barriers to the entry of bleaching chemicals [21].The advantages of using xylanase in biobleaching process include its application to different types of pulps, reduction in chlorine consumption for the bleaching sequence by up to 40%, decline in aromatic organic halides (AOX) in the bleach effluent by 25-30%, decreased ratio of COD/BOD in the effluent, reduction in colour of the effluent and attainment of higher-brightness ceilings. Enzymatic pretreatment of pulp with cellulase-free, thermo tolerant, and alkali stable xylanase produced by *Bacillus pumilus* SV-85S reduced the chlorine consumption by 29.16 %. Pre-bleaching with xylanase also led to improvement in various pulp strength properties like tensile strength, breaking length, burst factor, and tearness [22].

In addition, the use of xylanase helps in increasing pulp fibrillation, reduce beating times in the original pulp and increase the freeness in recycled fibres. Thus, the use of xylanase in pulp and paper industry causes a decrease in the consumption of chlorine for pulp bleaching thereby reducing the discharge of toxic organochlorine compounds into the effluent and consequently, lowering environmental pollution impact [22]. In other words, biobleaching with xylanase is an eco-friendly process. Furthermore, the reduced need for chlorine-based bleaching chemicals may decrease the cost of production. Sridevi et al., [23] investigated the bleaching efficiency of a crude xylanase for the recycling of wastepaper pulp and a significant reduction in kappa number and increase in brightness in enzyme-treated paper pulp than - untreated pulp were observed.

Desirable properties of xylanase for biobleaching

The desirable properties of xylanase as a bleach booster are: cellulase-free in nature, stable on kraft-pulp, active at alkaline pH, good thermal stability, cost-effective production and low molecular weight. The use of cellulase-free xylanase in pulp and paper industries selectively removes hemicellulose components with minimal damage to cellulose. Alkaline and thermostable xylanases are required as pulping and bleaching are both performed at high temperature and alkaline pH. Low molecular weight xylanase will facilitate better penetration in lignocellulosic complex. In order to make the process economically sound, low-cost production of xylanase is desirable.

Xylanase production

Xylanase is produced by a variety of microorganisms, including bacteria, fungi, and actinomycetes [24]. The first xylanase produced was partially purified from *Aspergillus foetidus* about half a century ago [25]. Bacteria are preferred over fungi as they are good source of alkaline and thermostable enzymes. Initially xylanases from fungal isolates were used in pulping process, but later due to various factors such as presence of cellulase, less stability of enzyme and low optimum pH these were found limited acceptability on commercial scale. It would be advantageous to use xylanase from bacterial sources as pre bleaching agents [26]. *Bacillus* species have been exploited as good xylanase producers. Xylanases can be produced in high titres using inexpensive and abundantly available agro-residues as substrates in both solid state (which involves the microbial growth & fermentation on moist, water insoluble & solid substrate in absence or near absence of free water) and submerged fermentation (which involves the microbial growth & fermentation in the presence of sufficient water so as to dissolve the whole medium component in it). Solid state fermentation is more economical than submerged fermentation mainly due to the cheap and abundant availability of agricultural wastes which can be used as substrates. Besides, SSF offers distinct advantages over submerged fermentation including economy of space needed for fermentation; simplicity of the fermentation media; no requirement for complex machinery, equipment and control systems, superior yield etc. [27]. The production of xylanases at low cost using cost-effective substrates has been reported by various workers [28-30].

Conclusion

Urged by environmental and legislative pressure together with advanced techniques and commitment from industry leaders, the pulp and paper industry has substantially reduced its environmental impacts on air, water and land over recent decades. In addition, biobleaching with xylanase may be cost-effective as it reduced the consumption of chlorine required for bleaching by up to 40%. The importance of enzymes is likely to increase as consumers demand more eco-friendly processes / products free of chemical additives.

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