

Chapter 13

Recent Advances in Physico-chemical and Biological Techniques for the Management of Pulp and Paper Mill Waste



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Abstract Pulp and paper industries are one of the major sources of environmental pollution that discharge enormous amount of wastewaters containing recalcitrant pollutants into the environment. Wastewaters have high biological oxygen demand (BOD), chemical oxygen demand (COD), total solids (TS), phenols, lignin and its derivatives. High strength of wastewaters containing dark colour and toxic compounds from pulp paper industries causes serious aquatic and soil pollution. On terrestrial region, pulp and paper mill wastewater at high concentration reduces the soil texture and inhibits seed germination, growth and depletion of vegetation, while in aquatic system, it blocks the photosynthesis and decreases the dissolved oxygen (DO) level which affects both flora and fauna and causes toxicity to aquatic ecosystem. The high pollution load from pulp and paper industrial wastewater gradually increases, and hence, there is a need for adequate treatment to reduce these pollution parameters before final discharge into the environment. Thus, this chapter gives detailed information about sources, characteristics, toxicity and physico-chemical and biological methods for the treatment of pulp and paper mill wastes and wastewaters.

Keywords Pulp and paper mill wastewater · Recalcitrant pollutants · Bioremediation

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1 Introduction

Industries such as pulp and paper, distillery, tannery and textile are one of the major sources of serious environmental pollution (Bharagava and Mishra 2017). Pulp and paper (P&P) mills are categorized as a core industrial sector and rank third in the world after primary metals and chemical industries in terms of freshwater used (Thompson et al. 2001; Sumathi and Hung 2006; Asghar et al. 2008; Mishra and Bharagava 2016). The pulp and paper industry is one of the most important industries of the North American and US economy (Nemerow and Dasgupta 1991). In terms of production and total earning, pulp and paper industries of Canada play a major role in country's economy, and estimated 50% of pulp and paper wastes are dumped into Canada's waters (Sinclair 1990). At present, more than 759 pulp and paper industries are present in India. The Indian pulp and paper industries are highly water intensive, consuming 100–250 m³ freshwater/ton paper (Singh. 2004) and generating a subsequent 75–225 m³ wastewater/ton paper (Ansari 2004). Out of 759 mills, 114 (15%) are large, 303 (40%) are medium and 342 (45%) are small. The pulp and paper industry is divided into the three sectors based on raw materials used: wood and bamboo mills using wood and bamboo produce 3.19 million tonnes, which is 31% of the production, and there are 26 large integrated paper mills. There are 150 agro-based mills using agro-residues like bagasse, wheat and rice straw, etc. that produce 2.2 million tonnes, which is 22% of the total production. Recycled fibre mills using wastepaper contribute almost 47% of the country's current production which is 4.72 million tonnes, and there are 538 recycled paper mills in operation. The environmental effects of agro-residue-based paper mills are of particular concern as these units generate 150–200 m³ effluent/ton paper with a high pollution load of 90–240 kg suspended solid (SS), 85–370 kg biochemical oxygen demand (BOD) and 500–1100 kg chemical oxygen demand (COD)/ton paper (Mathur et al. 2004). Indian paper mills are highly fragmented on the basis of production, i.e. printing and writing is 38.58%, packaging is 53.61%, while newsprint is 7.81%. According to the Ministry of Environment and Forest, Government of India, the pulp and paper sector is in the “Red Category” list of 17 industries having high polluting potential due to its serious pollution menace, and it is compulsory for pulp and paper mills to follow the appropriate standards set by Central Pollution Control Board (CPCB 2001). Wastewaters of pulp and paper mills impart a dark brown/black colour to the receiving water bodies. Lignin and chlorinated organic compounds are the key environmental pollutants released from pulp and paper industry. The offensive colour of these wastewaters principally of lignin and its degradation products produced during manufacturing process of papermaking reduces the transmission of light in waterways, reduces the aquatic plant photosynthesis and dissolved oxygen (DO) content and ultimately causes the death and putrefaction of aquatic fauna (Sahoo and Gupta 2005; Karrasch et al. 2006; Singh et al. 2016). On terrestrial environment, accumulation of toxic pollutants and metals in soil affects growth and development of plants. Chlorinated compounds released from pulp and paper mills are highly toxic and have carcinogenic, clastogenic, mutagenic and

endocrine effects on receiving bodies. Various extractives are also used in pulp and paper manufacturing process, although the concentration of these extractives is present in trace amounts, but some of them are very harmful for environment. Pulp and paper mill wastewaters pollute all aspects of life, i.e. water, soil and air, causing a major threat to the environment. In many developing countries like India, farmers irrigate their fields by wastewaters, which are released by pulp and paper mills, having high levels of several toxic compounds. Hence, the adequate treatment is necessary of these pollutants before the final discharge into the environment. Although several physico-chemical methods (precipitation, sorption, ozonation, ultrafiltration, reverse osmosis and electrochemical treatment) or combination of different methods are available for the treatment of pulp and paper mill wastewater, these methods are more energy intensive, produce secondary sludge and are cost-ineffective (Singhal and Thakur 2009a, b; Raj et al. 2014). The biological treatment is known to be very effective in reducing the organic load and toxic effects of kraft mill wastewaters (Park et al. 2007). Various wood extractives present in wastewaters are removed by biological methods. This treatment also reduces colour, COD, BOD and low-molecular-weight chloro-lignins (Nagarthamma et al. 1999; Barton et al. 1996). Microorganisms treat the wastewaters generally by the action of enzymes. The various enzymes involved in the treatment of pulp and paper mill wastewaters are lignin peroxidase, manganese peroxidase, laccase, etc. Microorganisms showing good production of these enzymes have the high potential to treat the wastewaters (Hooda et al. 2015; Kumar et al. 2016).

2 Sources and Characteristics of Pulp and Paper Wastewater

Wood is the most abundant source of pulping and papermaking process; this process consists of several toxic compounds which are washed away from the pulp fibres during the washing, dewatering and screening processes. Pulp fibres can be prepared from a vast majority of plants such as woods, straws, grasses, bamboos, canes and reeds. Among various stages of paper manufacturing process, the most considerable sources of pollution from pulp and paper mills are wood preparation, pulping, pulp washing and bleaching. Along with these processes, pulping stage generates a high-strength wastewater called black liquor which is dark brown in colour due to dissolved lignin and its degradation products, hemicelluloses, resin acids, unsaturated fatty acids, etc. (Berryman et al. 2004). About 200 m³/tonne of pulp is produced during pulping process, and it generates a huge amount of wastewaters (Cecen et al. 1992) which are highly polluted that cannot be easily recovered. During bleaching process, various types of phenolic compounds are produced; lignin and its derivatives are the main source of these compounds (Amat et al. 2005). Pentachlorophenol is generated unintentionally in wastewaters as a byproduct, and other most toxic pollutants such as coloured compounds, chlorinated phenols, chlorinated dibenzo-p-dioxin and dibenzofuran, chlorinated hydrocarbon, etc. are also released. Wastewaters produced from papermaking process contain cellulose (fines)

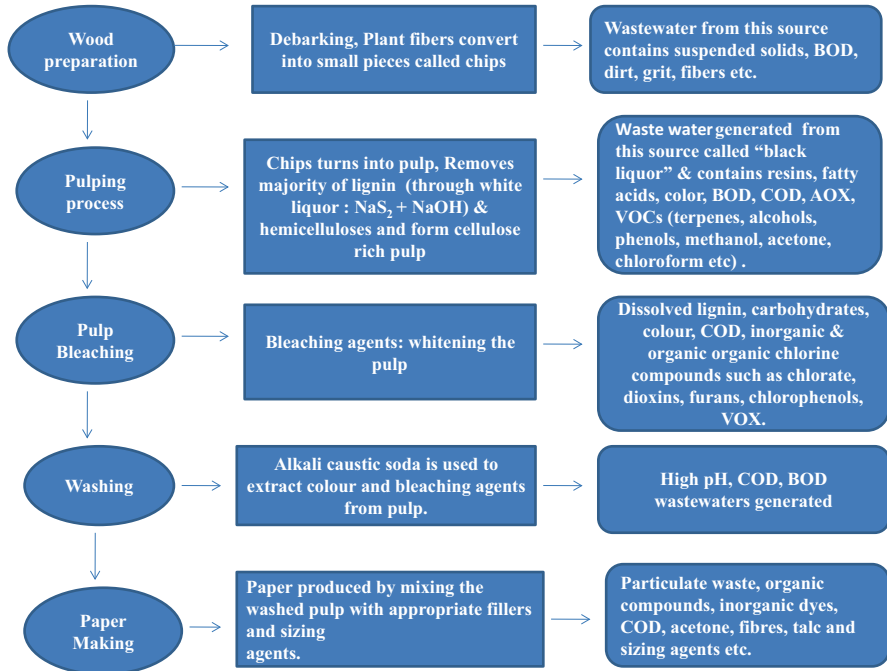


Fig. 13.1 Various stages and produced pollutants of pulp and paper mill wastewater

and other additives; this contaminated wastewater is referred as whitewater, and it can be up to 50% of the total mass. The pollutants released from various stages of the pulping and papermaking process are present in Fig. 13.1. Wastewaters produced from de-inking process contain ink residues, and sludges are also generated from wastewater treatment plant. These wastewaters are the significant concern for the environment because of highly toxic chlorinated compounds (EPA, 2002).

3 Effects of Pulp and Paper Wastewater Pollutants on Environment

Harmful pollutants released from pulp and paper industries affect all aspects of the environment: water, air and land (Makris and Banerjee 2002). Several studies confirmed the toxic and genotoxic effects of pulp and paper mill wastewaters on aquatic as well as on terrestrial environments. On aquatic fauna, the detrimental effects such as liver damage, oxidative and respiratory stress, mutagenic and genotoxic effects and other such lethal effects due to exposure of pulp and paper mill wastewaters

were reported by several authors (Owens et al. 1994; Ali and Sreekrishnan. 2001; Pokhrel and Viraraghavan. 2004; Kumari et al. 2016). Mandal and Bandana (1996) reported health impacts such as diarrhoea, vomiting, headaches, nausea and eye irritation on children and workers due to pulp and paper mill wastewater which is discharged into the environment. High level of carbon dioxide in pulp and paper mill wastewaters as a potential source of distress and toxicity to rainbow trout was reported by O'connor et al. (2000). Yen et al. (1996) reported the possibility of sub-lethal effects on aquatic organisms of Dong Nai River in Vietnam due to the discharged wastewaters from pulp and paper industries. Baruah (1997) reported serious concerns on surface plankton population change in Elengabeel's wetland ecosystem in India, because of untreated paper mill wastewater discharged into the receiving system. Howe and Michael (1998) studied the harmful effects of the treated pulp mill wastewater on irrigated soil in northern Arizona, which showed serious change in soil chemistry. Dutta (1999) investigated the toxic effects of paper mill wastewater (treated) applied to paddy field in Assam, India. Gupta (1997) and Singh et al. (1996) reported high levels of organic pollutants derived from paper mill wastewater in Tamil Nadu and Punjab, India, respectively. Skipperud et al. (1998) and Holmbom et al. (1994) reported various trace metals present in pulp and paper mill wastewaters at low concentrations. Pulping process is the most significant potential for environmental pollution. In pulping countries, the effect of the generated wastewater on the environment can be much greater. Most studies found that the bleached and unbleached kraft pulp mill wastewaters cause impaired liver function and also demonstrated a variety of responses in fish population's exposure to bleached wastewaters of kraft mills in the USA and Canada. These include smaller gonads, delayed sexual maturity, depression in secondary sexual characteristics and changes in fish reproduction (Munkittrick et al. 1997). In secondary wastewater treatment plants in the UK, the main problem that occurs is the growth of sewage fungus (Webb 1985), and other problems arise when the treatment of pulp and paper mill wastewater failed due to released suspended solids; it loses the nutrients, such as nitrogen and phosphorus, which can lead to eutrophication in recipient bodies. In agriculture, pulp and paper mill wastewaters are used for irrigation purpose, and it affects not only the crop growth and soil properties but also the mobility of various ions present in soil which is beneficial to plants (Ugurlu et al. 2008; Kumar and Chopra 2012). Chlorinated organic compounds such as dioxins and furans are supposed to cause skin disorders including skin cancer and also show reproductive effects in exposed organisms (Nestmann 1985; Malik et al. 2009), and adsorbable organic halides (AOXs) may bioaccumulate in fish tissues causing a variety of clastogenic, carcinogenic, endocrine and mutagenic effects, which may then also create problems to humans after consumption of the contaminated fish. Chlorinated phenols are also responsible for toxicity to both flora and fauna. During biological treatment process, wood extractives (e.g. resin acids and sterols) can be transformed into other toxic compounds, and severe toxicity may occasionally occur.

4 Treatment Approaches for Pulp and Paper Wastewater

Pulp and paper mill wastewaters hold a number of toxic compounds which are harmful for receiving bodies, are recalcitrant to degradation and can potentially induce toxicity, generally at the reproductive level (Costigan et al. 2012; Hewitt et al. 2008; Waye et al. 2014). Various treatment technologies and its application are shown in Table 13.1 and Fig. 13.2. Pollution from pulp and paper industries can be reduced by various methods as follows:

4.1 *Physico-chemical Treatment Approaches*

Plenty of literatures are available on various physico-chemical treatment methods that include sedimentation, flotation, adsorption, coagulation, oxidation, ozonation, electrolysis, reverse osmosis, ultrafiltration and nanofiltration technologies to remove suspended solids (SS), colloidal particles, floating matters, colours and toxic compounds from the produced harmful wastewaters.

4.1.1 Sedimentation and Flotation

Primary clarification may be attained by either sedimentation or flotation. Suspended particles present in pulp and paper mill wastewaters primarily consist of bark particles, fibres, fillers and coating materials (Pokhrel and Viraraghavan 2004). Sedimentation was the preferred option in the UK and approximately 80% removal of the suspended solids (Thompson et al. 2001). Dissolved air flotation is a conventional method for the removal of suspended solids and has been widely used in treatment of various types of industrial wastewaters. Gubelt et al. (2000) reported that dissolved air flotation method removes 65–95% total suspended solids (TSS), and it was an unstable unit, while Wenta and Hartmen (2002) stated that dissolved air flotation was able to remove 95% of the TSS. Implementation of such these treatment methods may depend upon the employed pulp and paper production process as well as applied secondary treatment methods (Kamali and Khodaparast 2015).

4.1.2 Coagulation and Precipitation

In case of pulp and paper mill wastewater treatment, coagulation and flocculation methods are basically employed in the tertiary treatment systems but not used in primary treatments. The basic principle of such methods is addition of metal salts to the wastewater stream to generate larger flocs from small particles. Wang et al. (2011) used aluminium chloride as coagulant and a modified natural polymer (starch-g-PAM-g-PDMC) as flocculant for the treatment of wastewaters from

Table 13.1 Various treatment technologies for the remediation of pulp and paper mill wastewaters

Treatment approaches	Properties	Applications	References
<i>Physico-chemical treatments</i>			
Sedimentation and flotation	In sedimentation, the gravity is used to separate solid phase from liquid, while in flotation process, buoyancy is increased of solids by forming gas bubbles	Industrial (paper, food, oil and plastic industries) and domestic wastewater treatment	http://www.gunt.de/images/download/flotation_sedimentation_english.pdf
Coagulation and flocculation	In coagulation, particles aggregate with themselves by the influence of a change in pH, and in flocculation process, the particles aggregate by the use of polymers that bind them together	Removal of organic matter, pathogen removal, removal of inorganics (arsenic and fluoride), all types of wastewater treatment	http://aquarden.com/technology/coagulation-and-flocculation . http://www.iwapublishing.com/news/coagulation-and-flocculation-water-and-wastewater-treatment
Electrochemical methods	They use electron as unique reagent	Colour removal in wastewater treatments and degradation of nonbiodegradable dyes do not produce solid waste residues	Sala and Gutierrez-Bouzan (2012)
Membrane technologies	It generates stable water without the addition of chemicals, low energy use, easy and well-arranged process	Efficient recovery of waste materials, impurities and byproducts from pulp and paper mill effluents prior to discharge. Treatment of industrial wastewaters especially treating wastewater from petrochemical and steel industry and power generation	Ebrahimi et al. (2016), Chen (2008), and Chen et al. (2009)
Adsorption	It is a surface phenomenon. Nature of the bonding between adsorbate and adsorbent: physic sorption (weak van der Waals forces), chemisorption (covalent bonding) and electrostatic attraction	Remove inorganic and organic pollutants, i.e. persistent organic pollutants (POPs)	Rashed (2013)

(continued)

Table 13.1 (continued)

Treatment approaches	Properties	Applications	References
Advanced oxidation process	In this process, the main mechanism is the generation of highly reactive free radicals	Ground remediation, removal of pesticides from drinking water, removal of formaldehyde and phenol and reduction in COD from industrial wastewaters	Ayed et al. (2017)
<i>Biological treatments</i>			
Activated sludge process	Complex mixture of microbiology and biochemistry	Activated sludge process has been the major treatment method for pulp and paper mill effluents in recent years. It removes organic substances (AOX), nutrients, BOD, COD as well as toxic compounds and pathogens from produced wastewaters	Ashrafi et al. (2015) and Wells et al. (2011)
Aerated lagoons		Efficient removal in BOD (over 95%) and chlorinated phenolics (85%) from pulp and paper mill wastewaters and other industrial wastewaters	Kamali and Khodaparast (2015)
Anaerobic digestion	Biochemical reactions occur and convert organic polymers from the feedstock into methane (biogas) and nutrient-rich digestate. It works best at temperatures of 30–60 °C	Used for reducing excess sludge volumes, energy efficient with lower biomass production	Garg and Tripathi (2011) and Metcalf and Eddy et al. (2003)
Bacterial treatment	Bacteria show enhanced biodegradation capability, mainly due to the broad pH range tolerability, biochemical versatility and immense environmental adaptability	Removes all types of pollutants from industrial wastes and wastewaters	Chandra and Singh (2012)
Fungal treatment	Produces extracellular enzymes and can survive at higher effluent load	Degradation of lignin/phenolic compounds from pulp and paper mill wastewaters and also reduction in colour, BOD, COD and AOX from different industrial wastewaters (textile, leather, distillery etc.)	Sankaran et al. (2010)

Algal treatment	Main mechanism for lignin removal by algae is metabolism rather than adsorption, and the main mechanism of colour and organic removal is partially metabolism and partially transformation	Treats several types of industrial wastewaters and also removes metals from wastewaters	Tarlan et al. (2002) and Usha et al. (2016)
Upflow anaerobic sludge blanket reactor (UASB)	Forms agglomerates (0.5–2 mm in diameter), and gas formed causes sufficient agitation in the reactor	Treats various industrial wastewaters like petroleum, distillery, canning industry, heavy metals, paper and pulp, tannery, pharmaceutical, domestic wastewater, etc.	Kaviyarasan (2014)
Sequencing batch reactor (SBR)	Fills and draws activated sludge system for wastewater treatment, uniquely suited for wastewater treatment applications characterized by low or intermittent flow conditions	Treats industrial wastewater containing phenolic compounds, such as p-nitrophenol (PNP) which is a hazardous chemical widely used in agricultural, pharmaceutical and dye industries as a synthetic intermediate in the manufacturing process and also treats both municipal and industrial wastewaters including dairy, pulp and paper, tanneries and textiles	Dutta and Sarkar (2015)
Constructed wetlands	Use natural functions of vegetation, soil and organisms to treat different water streams	They are capable of removing nutrients, biochemical oxygen demand, chemical oxygen demand, total suspended solids, colour, metals and toxic compounds from industrial wastewaters of different origin	Chaudhary et al. (2011)
Enzymatic treatments	Remove pollutants by precipitation or transformation to other value-added products	Application to biorefractory compounds; operation at high and low contaminant concentrations; work over a wide range of pH, temperature and salinity; absence of shock loading effects; reduction in sludge volume (no biomass generated) and the ease of controlling the process. Treat phenolic contaminants and related compounds, pulp and paper wastes, pesticides, cyanide wastes, food-processing wastes, removal of heavy metals, surfactant, oil and grease degradation	Karam and Nicell (1997)

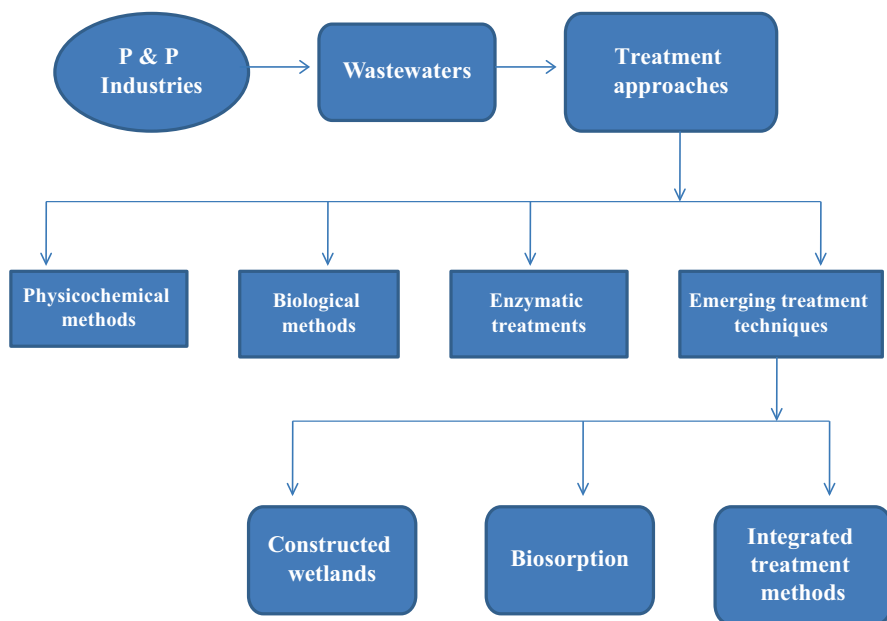


Fig. 13.2 Different methods used in the treatment of pulp and paper mill wastewater

primary sedimentation tank and concluded that at optimum condition, turbidity, lignin removal efficiency and water recovery were 95.7%, 83.4% and 72.7%. Tong et al. (1999) and Ganjidoust et al. (1997) have done a comparative study between horseradish peroxide (chitosan) and other coagulants such as $\text{Al}_2(\text{SO}_4)_3$, polyethyleneimine (PEI) and hexamethylene diamine epichlorohydrin polycondensate (HE), to remove colour, total organic carbon (TOC) and adsorbable organic halides (AOX), and observed that chitosan was a more better coagulant in removing these pollutants than others. Dilek and Gokcay (1994) reported that by using alum as coagulant, 96% removal of chemical oxygen demand (COD) from papermaking process, 50% from pulping stage and 20% from bleaching wastewaters were attained. Rohella et al. (2001) stated that polyelectrolytes were more effective than the conventional coagulant alum for removal of colour, turbidity and COD. Eskelinen et al. (2010) found that by using chemical precipitation method using 5 g/L of CaO, up to 90% removal of chemical oxygen demand (COD) is attained.

4.1.3 Electrochemical Methods

There has been an increasing attention towards electrochemical techniques (Malkin, 2002), which is an attractive, alternative and eco-friendly process for treating wastewaters in large scale (Soloman et al. 2009). This process includes electrocoagulation and electrooxidation. Different types of technical problems arise in pulp and

paper industries, which have been solved by electrochemical method. Electrochemical technologies are of great attention because of their more versatility and environmental compatibility, which makes the treatment of any type of pollutants (liquids, gases and solids) possible. In electrochemical methods, chemicals are not required; the main reagent is the electron, which is a clean reagent (Inan et al. 2004). Chanworrawoot and Hunsom (2012) found that the electrochemical method was much efficient to reduce the pollutants of various types of industries and produce low-density sludge in a very small amount. They also assist the reduction of lignin as well as organic and inorganic compounds, colour, total biological oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS) and total dissolved solids (TSD). In their batch culture experiment, the removal of colour, BOD and COD were 98%, 98% and 97%, respectively.

4.1.4 Membrane Technologies

Membrane filtration is a separation process that employs semipermeable membrane to divide the supply wastewater stream into two parts: the first part is permeate that contains the material passing through the membranes and the second is retentate in which the species are being left behind (Mallevalle et al. 1996). Membrane filtration achieved the efficient recovery of waste materials, byproducts and several impurities discharged from wastewaters. Recently, various membrane technologies have been used for the treatment of pulp and paper mill wastewaters. Reverse osmosis is an important method having the ability to destroy the pathogens (Asano and Cotruvo 2004). Dube et al. (2000) reported that 88% and 89% removal of BOD and COD was attained by reverse osmosis. The most important applications of membrane filtration techniques are the recovery of lignosulfonate from spent sulphite liquor and lignin from kraft black liquor; it also saves energy and is beneficial for the environment (Olsen 1980). Ciputra et al. (2010) found that nanofiltration technique removes 91% dissolved organic carbon from biologically treated newsprint mill wastewater. Gonder et al. (2011) revealed that at optimized conditions, membrane fouling can be minimized by nanofiltration process. They also investigate by using ultrafiltration membrane method the treatment of pulp and paper mill wastewater which occurs, and they achieved 83%, 97%, 95%, 89% and 50% removal of total hardness, sulphate, spectral absorption coefficient, COD and conductivity, respectively. Krawczyk et al. (2013) recovered high-molecular-mass hemicelluloses, from chemical thermo-mechanical pulping process wastewater, by using a membrane filtration.

4.1.5 Adsorption

Adsorption is one of the most prominent methods for the treatment and removal of inorganic and organic pollutants present in pulp and paper mill wastewaters. Adsorption has many advantages over other conventional methods like simple

design, low cost and land requirement. Recently, the low-cost adsorbents such as agricultural wastes, natural wastes and industrial wastes that have pollutant-binding capacities are used and are easily available. Activated carbon can be used as adsorbent for the treatment of water and wastewaters which is produced from these materials (Crini 2005). Various adsorbents such as coal ash, silica, fuller's earth, activated carbon, etc. have revealed adequate results for decolourization and pollutant removals from pulp and paper mill wastewaters, as reviewed by Pokhrel and Viraraghavan (2004). Ciputra et al. (2010) investigated the adsorption mechanism of granular activated carbon and ion exchange resin preferentially acts on the hydrophobic and high-molecular-weight fractions and reported 72% and 76% reductions in dissolved organic carbon by using this method. Das and Patnaik (2000) studied the removal efficiency of lignin from blast furnace dust (BFD) and slag by the adsorption mechanism was 80.4% and 61%. Shawwa et al. (2001) used activated coke as an adsorbent that removes 90% of colour, COD and AOX from bleached wastewater through adsorption process. Xilei et al. (2010) performed the adsorption method (low-cost bentonite as adsorbent) followed by the coagulation tertiary treatment (polyaluminum silicate chloride as coagulant), and they achieved 60.87% and 41.38% removals of COD and colour, respectively, at optimum doses of adsorbent and coagulant, i.e. 450 mg/l and 400 mg/l.

4.1.6 Oxidation

Advanced oxidation process (AOP) is the potential method for the remediation of wastewaters by conversion (oxidation) of complex recalcitrant compounds into inorganic substances (CO_2 and H_2O_2) or partial mineralization and transform them into less toxic forms. Generation of highly reactive free radicals is the main mechanism of advanced oxidation process. Hydroxyl radicals (OH^\bullet) are effectively destroying organic pollutants through the action of electrophiles that react rapidly with all electro-rich organic compounds (Covinich et al. 2014). Perez et al. (2002) proved that the combinations of fenton and photo-fenton reaction methods are highly effective for the treatment of bleached kraft mill wastewaters. Sevimli (2005) compared ozonation and combination of ozonation with H_2O_2 oxidation and fenton oxidation for the removal of COD and colour from pulp and paper mill wastewaters and analysed that ozonation and ozonation with hydrogen peroxide successfully remove the colour, while Fenton's oxidation process was more effective in reducing the COD and colour. Ozonation process is also used to oxidize chemicals such as guaiacol, syringaldehyde, vanillin, phenol, trichlorophenol, chlorophenol and cinnamic acid derivatives, which present in pulp and paper mill wastewaters (Fontanier et al. 2005; Hermosilla et al. 2014). Acid orange 7 is a typical dye used in pulp and paper industries, which is degraded by heterogeneous catalytic wet hydrogen peroxide process (Herney-ramirez et al. 2011).

5 Biological Treatment Approaches

Biological methods in which microorganisms such as fungi, bacteria and algae and their enzymes are used, as singly applied or in combination with physical and chemical methods to treat pulp and paper mill wastewaters (Singhal and Thakur 2009a, b). Most of the conventional treatment methods are not very effective for the removal of colour and degradation of recalcitrant compounds such as lignin (Balcioglu et al. 2007), but compared with physico-chemical methods, biological treatment methods are suitable to reduce COD, BOD and lignin from various types of pulp and paper mill wastewaters (Tiku et al. 2010). Detailed list of microorganisms for the treatment of pulp and paper mill wastewater is given in Table 13.2. Biological processes are divided into two categories, namely, aerobic and anaerobic, and it depends on the type of microorganisms.

5.1 Aerobic Process

Aerobic treatment processes such as activated sludge (AS) and aerated lagoons (AL) are the commonly used treatment methods for pulp and paper mill wastewaters. Activated sludge system is the major treatment method capable of removing huge amount of sludge and secondary pollutants generated from pulp and paper industries (Buyukkamaci and Koken 2010). Lots of literature have been published for the treatment of pulp and paper wastewaters through activated sludge system. Chandra (2001) reported the efficient removal of colour, BOD, COD, phenols and sulphide by microorganisms such as *Pseudomonas putida*, *Citrobacter* sp. and *Enterobacter* sp. through activated sludge process. Bengtsson et al. (2008) treated pulp and paper mill wastewaters from recycled fibres by activated sludge system and found 95% removal of COD. Mahmood and Paice (2006) and Ghoreishi and Haghighi (2007) treated various types of pulp and paper wastewaters through aerated stabilization basin and remove 50–70% BOD, 30–40% COD, AOX and chlorinated compounds. Aerated lagoon is the simple and cost-effective biological system which is relevant on both lab scale as well as full scale for pulp and paper mill wastewaters. This system is used to remove BOD, low-molecular-weight AOX and fatty acids at full-scale applications (Bajpai 2001). Schnell et al. (2000) reported that the aerated lagoon system removes BOD, AOX and phenols. Pokhrel and Viraraghavan (2004) reviewed that aerated lagoon system was efficient in removal of chlorinated phenols (85%) and BOD (>95%) from pulp and paper mill wasters.

Table 13.2 Microorganisms involved in the treatment of pulp and paper mill wastewater

Microorganisms	Used for the removal of pollutants	References
Fungal sp.		
<i>Trametes pubescens</i>	Chlorophenols	Gonzalez et al. (2010)
<i>Aspergillus niger</i>	Alkaline peroxide mechanical pulping effluent	Liu et al. (2011)
<i>Emericella nidulans var. nidulans</i>	Colour and lignin	Singhal and Thakur (2009a, b)
<i>Phanerochaete chrysosporium</i>	Colour, lignin and COD	Saritha et al. (2010)
<i>Cryptococcus</i> sp.	Colour, lignin and toxicity of the effluent	Singhal and Thakur (2009a, b)
<i>Trichoderma</i> sp.	Colour	Saravanan and Sreekrishnan (2005)
<i>Aspergillus flavus</i> F10	Colour and lignin	Barapatre and Jha (2016)
<i>Fibrodontia</i> sp. RCK783S	Colour	Kreetachat et al. (2016)
<i>Rhizopus arrhizus</i>	Lignin and chlorophenols	Lokeshwari et al. (2015)
Bacterial sp.		
<i>Pseudomonas fluorescens</i>	Colour, lignin, COD, phenol, chloride content	Chauhan and Thakur (2002)
<i>Paenibacillus</i> sp.	Colour, lignin, BOD, COD, phenol	Raj et al. (2014)
<i>Citrobacter freundii</i> , <i>Serratia marcescens</i>	TOC, COD, lignin	Abhishek et al. (2015)
<i>Alcaligenes faecalis</i> and <i>Bacillus cereus</i>	COD	Mehta et al. (2014)
<i>Bacillus subtilis</i> subsp. <i>inaquosorum</i>	Lignin, colour, COD	Hooda et al. (2016)
<i>Citrobacter freundii</i> and <i>Citrobacter</i> sp.	COD, AOX, colour, lignin	Chandra and Abhishek (2010)
<i>Paenibacillus glucanolyticus</i>	Deconstruct pulping waste	Methews et al. (2014)
<i>Klebsiella</i> sp., <i>Alcaligenes</i> sp. and <i>Cronobacter</i> sp.	Colour, AOX, TDS, TSS	Kumar et al. (2014)
Algal sp.		
<i>Scenedesmus</i> species	Nutrients, organic pollutants, BOD, COD	Usha et al. (2016)
Microalgae	Convert secondary waste into value added products	Kouhia et al. (2015)
<i>Chlorella</i>	COD, colour, AOX, chlorinated compounds	Tarlan et al. (2002)

5.2 Anaerobic Process

Anaerobic process is considered to be more suitable method for the treatment of high-strength organic wastewaters. Lots of literature have been published on anaerobic process along with microbial communities to treat pulp and paper mill wastewaters (Ince et al. 2007). In recent years, a stable biological process, anaerobic digestion (AD), is used for treatment of high loads of pulp and paper mill wastewaters. This approach has several advantages over various conventional treatment methods such as simple design, reduction of produced sludge volume up to 30–70%, destruction of pathogens in the thermophilic region, cost-effective and eco-friendly in nature (Zwain et al. 2013; Ekstrand et al. 2013). The anaerobic treatment of pulp and paper mill wastewater results in the degradation of pollutants such as lignin and their derivatives, fatty acids, resins and organic compounds, which are produced during the various steps of papermaking process (Sumathi and Hung 2006).

5.3 Bacterial Treatment

Several species of bacteria have been used for the remediation of pulp and paper mill wastewaters, and few of them have also been used commercially. Various studies have reported that some bacterial species (anaerobic and aerobic both) could metabolize lignin and their related compounds to low-molecular-weight compounds, and this is due to huge adaptableness and biochemical versatility of bacterial species (Chandra et al. 2007; Abhishek et al. 2015). Chandra et al. (2009) found that *B. cereus* and *Serratia marcescens* was capable to reduce colour (45–52%), lignin (30–42%), BOD (40–70%), COD (50–60%), total phenol (32–40%), and PCP (85–90%) in axenic conditions, but in mixed culture conditions, the reduction in colour, lignin, BOD, COD, total phenol and PCP recorded was 62%, 54%, 70%, 90%, 90%, and 100%, respectively. Tyagi et al. (2014) isolated two bacterial strains, *Bacillus subtilis* and *Micrococcus luteus*, and one fungi, *Phanerochaete chrysosporium*, from pulp and paper mill wastewaters and sludge; these microbes were capable in reducing BOD up to 87.2%, COD 94.7% and lignin content 97% after 9 days; pH was down to neutral, and dissolved oxygen increased from 0.8 to 6.8 mg/L. Further in recent studies, Raj et al. (2014) found that bacterial strain, i.e. *Paenibacillus* sp., effectively reduced colour (68%), lignin (54%), phenol (86%), BOD (83%) and COD (78%). Simultaneously, in this study, the toxicity with treated and untreated wastewater on the growth and germination of mung bean (*Vigna radiata* L.) seed was also performed (Kumari et al. 2014). Hooda et al. (2015) studied the degradation of pulp and paper mill wastewater by a rod-shaped Gram-positive bacterium, i.e. *Brevibacillus agri*, in batch culture and in semi-continuous reactor. During batch study, this bacterium reduced COD up to 69%, colour 47%, lignin 37% and AOX 39%, while in semi-continuous reactor study, it reduced COD up to 62%, colour 37%, lignin 30% and AOX 40%.

5.4 Fungal Treatment (Mycoremediation)

Fungi are common in the treatment of pulp and paper mill wastewaters (Yang et al. 2011). In comparison with bacteria, fungi can survive at higher strengths of wastewaters, and like bacteria, they also produce extracellular enzymes (Singhal and Thakur 2009a, b). Malaviya and Rathore (2007) stated the bioremediation of pollutants from pulp and paper mill wastewater by a novel consortium of white-rot and soft-rot fungi which reduced the colour, lignin and COD by 78.6%, 79% and 89.4%. It has been shown that wood-degrading white-rot fungus is very effective for the degradation of lignin and chlorinated compounds, which are mainly responsible for colour and toxicity of pulp and paper mill wastewaters (Saritha et al. 2010). White-rot fungus *T. pubescens* along with TiO₂/UV was used for degradation of chlorophenols, and this combination (biological and advanced oxidation process) allowed up to 100% chlorophenol removal (González et al. 2010).

5.5 Algal Treatment (Phycoremediation)

Algae are important bioremediation agents, and they used natural mechanism for the treatment of wastewaters. Tarlan et al. (2002) were found to remove 58% COD, 84% colour and 80% AOX from pulp and paper wastewaters by algae and also showed that they grew mixotrophically and partially metabolized colour and organic compounds (released from pulping stage) to non-coloured and simple molecules. Several studies have reported wastewater can be used for the cultivation of microalgae (Ramanna et al. 2014). Microalgal cultivation from wastewater has a twin purpose: supply nutrients and minimize the freshwater requirements along with the removal of COD and BOD from wastewaters. Algae can use huge amounts of organic compounds from wastewater for rapid growth in the photoheterotrophic or mixotrophic environment in the presence of light (Li et al. 2011). Usha et al. (2016) stated that microalgal treatment is an efficient tool for the remediation of pulp and paper industry wastewater, and in their lab study, they found maximum removal of BOD (82%) and COD (75%), respectively, through microalgal cultivation in outdoor open pond. Algae have also been used in the removal of heavy metals from wastewaters.

6 Biological Reactors Study Used in the Treatment of Pulp and Paper Mill Wastewaters

Various types of reactors/digesters including SBR, MBR, UASB, etc. are reported for the degradation and treatment of noxious and deleterious pollutant present in pulp and paper mill wastewater. These reactors are principally based on the anaerobic microbial treatment technology that can efficiently reduce high concentration of

pollution load and signifies wastewater quality for its reuse in irrigation and other practices. Khan et al. (2011) observed 87% removal of COD, and the turbidity removal was 95% from pulp and paper mill wastewaters through column-type sequencing batch reactor. The alkalinity and pH of the treated wastewaters were in the permissible range and improved the characteristics of produced sludge. Kumar and Subramanian (2014) found through sequential batch reactor (SBR) system the removal efficiencies of COD, BOD, TDS, TSS and organic compounds reached up to 84%, 83%, 85%, 88% and $80 \pm 4.5\%$ under the retention time of 24 h. Various studies showed that high loads of organic pollutants present in pulp and paper mill wastewater reduced through SBR (Milet and Duff 1996). Muhamad et al. (2013) reported a significant reduction in the pollutants from recovered fibres of pulp and paper mill wastewater by granular activated carbon-sequencing batch biofilm reactor (GAC-SBBR) and also achieved 97.2% removal of COD, 99.4% of $\text{NH}_3\text{-N}$ and 100% of DCP. Buyukkamaci and Koken (2010) stated that the activated sludge process is the most important treatment method for the removal of low and medium strength of pulp and paper mill wastewaters but has some drawbacks, which can be improved in combination with membrane bioreactors. Membrane fouling in membrane technologies can increase the maintenance and operational costs, which may also overcome by membrane bioreactors (Le-clech et al. 2006). Removal of COD by moving bed biofilm reactor (MBBR) and the amount of sludge which is produced in secondary treatment from pulp and paper industries can also be reduced by membrane bioreactors (Jahren et al. 2002). Upflow anaerobic sludge blanket (UASB) reactor also known as anaerobic reactor was used in the treatment of various industrial wastewaters like tannery, distillery, pharmaceutical, pulp and paper, etc. Microorganisms living in the sludge blanket of UASB having microbial granules of size 0.5–2 mm break down organic matter by anaerobic digestion into simple compounds and biogas and can be used as energy source. Buyukkamaci and Koken (2010) showed that upflow anaerobic sludge blanket reactor followed by an aeration basin is the most economic and technically feasible treatment for medium and high strength wastewaters. In previous studies, Peerbhoi (2000) investigated that an upflow anaerobic sludge blanket (UASB) reactor was not feasible, as the pollutants were not properly degraded.

7 Other Emerging Treatment Approaches

7.1 *Constructed Wetlands (CWs)*

Constructed wetlands (CW) are emerging, low-cost and eco-friendly sustainable wastewater treatment systems. These are engineered integrated wastewater treatment systems of plants, water, microorganisms and the environment. Plants, soil, sand and gravels make shallow beds or channels, and a variety of microorganisms grow on these beds or channels to improve the quality of wastewaters (USEPA

2004). Both systems are able to remove high biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), colour, metals and toxic compounds, which are present in various types of industrial wastewaters. Plants directly uptake the nutrients from soil and facilitate indirect aerobic degradation of pollutants. Conventional treatment methods are not very much capable of removing high organic pollutants and colour from pulp and paper mill wastewaters. Constructed wetlands should be a better treatment way for pulp and paper mill wastewaters because their treatment efficiency is higher than conventional treatment methods (Choudhary et al. 2011).

7.2 *Biocomposting*

Biocomposting is one of the most valuable and green treatment processes for the mitigation of various harmful pollutants of industrial wastewaters. This method is also suitable for the treatment of wastes and sludges especially produced from paper fibres and organic materials. In biocomposting, wastes are dumped with microorganisms; humus like mater is produced which may be used in agriculture, house-plant greenhouse, etc. (Christmas 2002; Gea et al. 2005). Microorganisms converted organic materials into CO₂, humus and heat. The increased temperatures (thermophilic phase) in composts found that the rapid degradation of lignocelluloses occurs and is mainly degraded by thermophilic micro-fungi and actinomycetes (Tuomela et al. 2000).

7.3 *Enzymatic Treatments*

Several enzymes such as peroxidases, oxidoreductases, cellulolytic enzymes, cyanidase, proteases, amylases, etc. are used to treat industrial wastewaters. Ligninolytic group (laccase, MnP and LiP) of peroxidases from a variety of different sources have been reported to play an important role in waste and wastewater treatment (Chandra and Chowdhary 2015). Recently the demand of these enzymes (laccase, LiP, MnP) has increased gradually due to their prospective applications in the diverse biotechnological areas. Lignin peroxidase (LiP) and Mn peroxidase (MnP) were reported to be very efficient in decolourization of kraft pulp mill wastewaters (Moreira et al. 2003). Predominantly white-rot fungus and their specific enzymes are required for lignin degradation. *Phanerochaete chrysosporium* is one of the most important white-rot fungus and widely studied model for lignin degradation. Bacterial laccases are most considerable in the remediation of pollutants of industrial wastes. Laccases are also involved in the treatment of various industrial wastewaters such as pulp and paper, textile, tannery, distillery, etc. (Sangave and Pandit 2006; Chandra and Chowdhary 2015; Mani and Bharagava 2016; Bharagava et al. 2017; Chowdhary et al. 2017a, b).

7.4 Biosorption

Sorption is a process in which one substance is attached to another and bio means the involvement of living entities, i.e. biosorption is a physiochemical process that can be defined as the involvement of live entity like fungi or bacteria (adsorbent) and chemical or metal (adsorbate) leading to the removal of substances from solution through biological materials (Aksu 2002; Gadd 2009). Biosorption is used to treat wastewaters produced from various industrial sectors, and this treatment is considered clean, efficient, cost-effective and easy to operate (Saiano et al. 2005). Singhal et al. (2015) isolated a fungus, *Emericella nidulans*, was used for biosorption of colour of pulp and paper mill wastewaters, and after treatment the fungus turned dark brown in colour.

8 Challenges

Pulp and paper industrial sectors are facing many critical issues during the wastewater treatment and disposal process. In this study, the following challenges were noted:

- Pulp and paper industry produces large amount of wastewaters causing serious concerns for environment.
- In developing countries including India, lack of advanced treatment techniques and waste disposal are a serious concern.
- High values of physico-chemical parameters such as COD, BOD, TSS, TDS, etc. cause toxic effects in the aquatic system.
- Less involvement of government agencies with the industries.
- Degradation of complex compound lignin, which is a major pollutant in pulp and paper industry.
- Proper dumping of sludge, produced during the papermaking process.

Thus, there is an urgent need to solve these issues for sustainable development.

9 Conclusion

This chapter concluded the following points:

- Pulp and paper industries have high strength of wastewaters, i.e. it contains high COD, BOD, TSS, TDS etc., which causes hazardous effects on living entities.
- Untreated wastewater, when discharged into the green belt, disturbs the ecological balance of environment.
- Effective treatment and proper disposal are thus needed.

- Pulp and paper mill wastewater can also be used for the irrigation purpose with proper dilution.
- Biological approaches for the mitigation/removal of pulp and paper mill wastewater pollutants are gaining its momentum in the arena of wastewater treatment methods.
- Biological treatment methods are commonly used because these are the only treatment technique that produces very useful byproducts, which may be in large demand, and also it is a significant need for sustainable development of eco-friendly environment.
- Physico-chemical treatment strategies are more expensive/costly than biological treatment approaches for the capability of both colour and organic load reduction.
- On the basis of available literature on pulp and paper mill wastewater treatment, it seems that there is a need to address these problems and limitations in conventional treatment approaches and find out an effective solution. Hence, this chapter covers all the issues related to pulp and paper wastes/wastewaters and treatment technology for the sustainable development of environment.

Acknowledgement The University Grant Commission (UGC) to Ms. Surabhi Zainith for his Ph.D. work from UGC, Government of India (GOI), New Delhi, India, is duly acknowledged.

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