

WOLKITE UNIVERSITY



COLLEGE OF NATURAL AND COMPUTATIONAL SCIENCE

DEPARTMENT OF BIOTECHNOLOGY

REVIEW ON BIOREMEDIATION OF WASTE WATER IN PULP AND PAPER MILL EFFLUENT

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LIST OF ABBREVIATION

| | |
|----------|----------------------------|
| AOX..... | Adsorbable organic halides |
| BOD..... | Biochemical oxygen demand |
| COD..... | Chemical oxygen demand |
| EOP..... | End-of-pipe |
| C..... | Carbon |
| N..... | Nitrogen |
| H..... | Hydrogen |

SUMMARY

Pulp and paper industry is considered as one of the most polluting industry contributing 100 million kg of toxic pollutants that are being released every year in the environment and more than 260 chemicals have been identified in effluents, which are produced at different stages of paper making in pulp and paper industry. The pulp and paper industry is typically associated with pollution problems related to high BOD, COD, toxicity, AOX, color, suspended solids, lignin and its derivatives and chlorinated compounds. Although numerous studies have looked ways by various researchers to remove COD, BOD, color etc. of pulp and paper effluents, the problem still persists. Number of microorganisms including bacteria, fungi and algae has been implicated to degrade the xenobiotic compounds present in pulp and paper mill effluent. The aim of the current review is to overcome the limitations of waste water treatment method currently used in pulp and paper mill to be of cost benefit and to be able to fulfill the environmental protection requirements.

Key words: *Industrial wastewater, Bioremediation, pulp and paper mills*

1.INTRODUCTION

Industrialization improves the economic value of a nation but simultaneously it leads to the degradation of environment (Hossain and Rao, 2014; Raj *et al.*, 2014). Economic benefits of the pulp and paper industry have led it to be one of the most important industrial sections in the world. Pulp and paper industry is considered as one of the most polluting industry contributing 100 million kg of toxic pollutants that are being released every year in the environment (Dey *et al.*, 2013). Still now, pulp and paper mills are facing challenges with the management of the consequential pollutants, considering the environmental feedbacks and enduring legal requirements (Kamali and Khodaparast, 2015). The industry is the sixth largest polluter (after oil, cement, leather, textile and steel industries) discharging a variety of gaseous, liquid and solid wastes into the environment (Monte *et al.*, 2009). More than 260 chemicals have been identified present in paper and pulp mill effluents which are produced at different stages of paper making (Hawkins *et al.*, 2002). Generally, the pulp and paper industry has been considered to be a major consumer of natural resources (wood, water) and a significant contributor of pollutant discharges to the environment (Monte *et al.*, 2009).

The pulp and paper industry typically generates large quantities of wastewater which require proper treatment prior to discharge to the environment; otherwise it represents a significant environmental and economic problem (Prasongsuk *et al.*, 2009). One significant problem is the persistent dark brown colour due to lignin and its derivatives, such as chlorolignin in the released effluent discharged from the pulp bleaching process (Prasongsuk *et al.*, 2009). The effluent cause considerable damage to the receiving water if discharged untreated since, they have a high Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), chlorinated compounds measured as adsorbable organic halides (AOX), suspended solids mainly fibers ,fatty acids, tannins, resin acids, lignin and its derivatives, sulfur and sulfur compounds, etc. (Chandra *et al.*, 2007). Some of these pollutants are naturally occurring wood extractives like tannins, resin acids, stilbenes and lignin. It is well established that many of these contaminants are acute or even chronic toxins. Chlorinated organic compounds like dioxins and furans have the ability to bring genetic changes in exposed organisms. This has resulted in a growing concern about the potential

adverse effects of genotoxicants on aquatic biota and public health through the contamination of drinking water supplies, recreational water or edible organic species (Chandra *et al.*, 2007, 2009).

The increasing public awareness of the fate of these pollutants and stringent regulations established by the various authorities and agencies are forcing the industry to treat effluents to the required compliance level before discharging in to the environment (D'Souza *et al.*, 2006). The effluent coming from the pulp and paper industry can be minimized by various internal process changes and management measures through the available technology (Waye *et al.*, 2014).

Physicochemical treatment processes include removal of suspended solids, colloidal particles, floating matters, colors and toxic compounds by conventional and non-conventional methods like sedimentation, flotation, screening, adsorption, coagulation, oxidation, ozonation, electrolysis, reverse osmosis, ultra-filtration and nano-filtration technologies, the replacement of chlorine by hypochlorite, sorption on hypo and alum-sludge, activated carbon and all phenolic compound (Wang *et al.*, 2011). However, these processes are quite expensive and none are considered to be commercially viable and still the problem remains unsolved (Gutierrez *et al.*, 2006). Compared with physicochemical ways, biological methods for wastewater treatment are considered to be of cost benefit, eco-friendly and suitable for reduction of the BOD and COD from the effluents (Singhal and Thakur, 2009).

This article reviews the origins and effects of major pollutants present in pulp and paper mill effluents and the progress made in their reduction by using microorganisms such as bacteria, fungi and algae.

2.OBJECTIVES OF THE REVIEW

The reviewed in the present paper, have aimed to overcome the limitations of waste water treatment method currently used in pulp and paper mill to be of cost benefit and to be able to fulfill the environmental protection requirements.

3.SIGNIFICANCE OF REVIEW

This reviews implies that the origins and effects of major water pollutants present in pulp and paper mill effluents and the progress made in their reduction by using microorganisms. Biological methods for wastewater treatment are considered to be of cost benefit, eco-friendly and suitable for reduction of the BOD and COD from the effluents.

4. Process description and sources of pollution

Paper making includes five basic steps and each step can be carried out by a variety of methods.

Debarking converts the plant fiber into smaller pieces called chips and removes the bark. In this step, the nature of the raw material used, i.e. hard wood, softwood, agro residues, results in the transfer of tannins, resin acids, etc.

Pulping turns the chips into pulp. This process removes the majority of lignin and hemicellulose content from the raw material, resulting in a cellulose rich pulp (Ali and Sreekrishnan, 2001).

Bleaching is engaged on the brown pulp obtained after pulping in order to meet the desired colour dictated by product standards. Several bleaching agents, including chlorine, chlorine dioxide, hydrogen peroxide, oxygen, ozone, etc. may be used either singly or in combination. It is in this step that lignin, phenols, resin acids, etc. get chlorinated and transformed into highly toxic xenobiotics (Ali and Sreekrishnan, 2001).

Washing removes the bleaching agents from the pulp. Generally, an alkali caustic soda is used to extract color and bleaching agents from the pulp and hence, this process is also known as the alkali extraction stage (Ali and Sreekrishnan, 2001) .

Paper and paper products are finally produced by mixing the washed pulp with appropriate fillers (clay, titanium dioxide and calcium carbonate) and sizing agents like rosin and starch (Ali and Sreekrishnan, 2001).

5. Advance treatment of paper and pulp mill effluent

Ample studies have specified that pulp and paper mill effluents can potentially induce aquatic toxicity, especially at the reproductive level (Costigan *et al.*, 2012; Hewitt *et al.*, 2008; Waye *et al.*, 2014). The effluent coming from the pulp and paper industry can be minimized by various internal process changes and management measures through the available technology (Waye *et al.*, 2014).

5.1. Physicochemical treatment processes

The process removal of suspended solids, colloidal particles, floating matters, colors and toxic compounds by conventional and non-conventional methods like sedimentation, flotation, screening, adsorption, coagulation, oxidation, ozonation, electrolysis, reverse osmosis, ultra-filtration and nano-filtration technologies. However, these processes are quite expensive and none are considered to be commercially viable (Gutierrez *et al.*, 2006) and still the problem remains unsolved, since lignin undergoes a spatial rather than chemical change and thus persists albeit in a different form (Singh and Kaur, 2015).

5.2. Biological treatment

Biological treatment methods involve the utilization of microorganisms including fungi, bacteria, algae and enzyme. Compared with physicochemical ways, biological methods for wastewater treatment are considered to be of cost benefit, eco-friendly and suitable for reduction of the BOD and COD from the effluents. (Abd El-Rahim and Zaki, 2005). Bioremediation is a pollution control technology that uses biological systems to catalyze the degradation or transformation of various toxic chemicals to less- harmful forms.

Thus, bioremediation is employed for the treatment of various industrial effluents like paper and pulp mill effluent (Wu *et al.*, 2005; Abd El-Rahim and Zaki, 2005; Yang *et al.*, 2008).

5.2.1. Remediation with algae

It has been reported that some algae like *Microcystis spp.*, can decolourize diluted bleach kraft mill effluents (Iyovo *et al.*, 2010; Sharma *et al.*, 2014). The study was observed that pure and mixed algal cultures removed up to 70% of colour within 2 months of incubation. All cultures showed a similar colour reduction pattern consisting of a phase with declining rate. The decolorization was most corrective during the first 15-20 days of incubation and then gradually declined. The decolorization by algae is caused by metabolic transformation of coloured molecules to colourless molecules with limited assimilation or degradation of molecular entities (Chandra and Singh, 2012). Recently a mixed culture of algal including *Chlorella*, *Chlamydomonas*, *Microcystis*, etc. were used for removal AOX and colour and it was reported that there was nearly a 70% AOX reduction while, color was reduced by 80% in 30 days under continuous lighting conditions. Several analysis of alkaline extraction of algal biomass and material balance findings indicated that the main color removal mechanism was metabolism rather than adsorption (Sharma *et al.*, 2014; Chandra and Singh, 2012).

5.2. 2. Remediation with bacteria

Number of bacterial species has been assessed for their decolorization abilities and a few of them have also been used commercially. the dominant aboriginal microbes like *Bacillus subtilis* and *Micrococcus luteus* were found competent of reducing BOD up to 87.2%, COD up to 94.7% and lignin content up to 97% after 9 days under shaking conditions and brought down pH of raw Pulp and Paper Mills Effluent (PPME) to neutral and also enhance the Dissolved Oxygen (DO) present in effluent from 0.8-6.8 mg/ L (Tyagi *et al.*, 2014).

Two bacterial strains *Pseudomonas putida* and *Acinetobacter calcoaceticus* were studied for degradation of black liquor from a kraft pulp and paper mill in a continuous reactor. They were able to remove 70-80% of COD and lignin while, the colour removal efficiency was around 80% in 8 days (Murugesan, 2003; Abd El-Rahim and Zaki, 2005). *Melhylobacterium* exhibited a limited substrate range but was capable of removing significant amounts of AOX from both hardwood and softwood effluents.

By contrast, *Pseudomonas spp.* revealed a limited substrate range and poor to negligible reduction in AOX levels from both effluent types (Keharia and Madamwar, 2003).

The isolated bacteria like *Pseudomonas putida*, *Citrobacter spp.* and *Enterobacter spp.* can decolorized effluent up to 97% and also can reduced BOD, COD, phenolics and sulfide upto 96.63, 96.80, 96.92 and 96.67%, respectively within 24 h of growth and the heavy metals were removed up-to 82-99.80% (Keharia and Madamwar, 2003; Tiku et al., 2010).

The extracellular xylanase from *Bacillus stearothermophilus* T6 is a thermostable alkali tolerant enzyme was successfully used in a large-scale bio-bleaching that bleaches pulp optimally at pH 9 and 65°C temperature.

Streptomyces badius and *S. viridosporous* were able to use a commercial kraft lignin as sole carbon source which was characterized by Fourier transformed infra-red spectroscopy, amino acid analysis, elemental analysis for C, H, N and high performance size exclusion chromatography (Abd El-Rahim and Zaki, 2005; Chandra et al., 2011).

Factors effect on degradations processes

Effect of additional carbon source: Additional carbon source plays important role in degradation process. Number of studies established that the color removal ability increased by about 20-25% by the addition of nutrients like glucose and sucrose. (Tiku et al., 2010).

Mechanism of color removal: Adsorption is playing major role to contribute the removal of color. *P.aeruginosa* was taken out from the wastewater, after the decolorization experiment and analyzed by scanning electron microscopy. Scanning electron microscopy studies showed that the bacterial cells adhering to the colored material present in the wastewater exhibited some extracellular secretion visible on the cell surfaces which was absent in the control cells. The surfaces of the cells growing in the wastewater were not as smooth as seen in the control cells. This would mean that adsorption could be a major contributor to color removal. However, the extent of adsorption was not clear and further study is required (Tiku et al., 2010).

5.2.3 Remediation with fungi

The presence of fungi are quite natural in pulp and paper mill effluent as well as sludge (Yang et al., 2011). They yield extracellular enzymes and can survive at higher effluent load, compared with bacteria (Kamali and Khodaparast, 2015). The white- rot fungus *Schizophyllum commune*

can be decolourized the effluent which is coming from bagasse based pulp and paper mills. But, this *Schizophyllum commune* could not degrade lignin unless metabolizable carbon source was made available concurrently (Dashtban et al., 2010). Sucrose was found to be the best co-substrate for the breakdown of lignin (Kamali and Khodaparast, 2015).

The efficiency of treatment of effluent with *Schizophyllum commune* was highest at pH 4-5 and was further improved by intermittent aeration. *Schizophyllum commune* can be removed the colour of effluent by 90% and also reduced BOD and COD by 70 and 72% respectively under optimum conditions in 2 days incubation (Saritha et al., 2010; Kamali and Khodaparast, 2015). *Tinctoria borbonica* has been shown to decolourize the kraft waste liquor to a light yellow colour and near about 90-99% colour reduction was achieved after 4 days of cultivation (Abd ElRahim and Zaki, 2005).

One of the most efficient lignin degrading and decolorizing white rot fungus is *Phanerochaete chrysosporium* which is called as model white rot fungus because of its specialized ability to degrade the abundant aromatic polymer lignin, which has been studied in much detail by Senthilkumar et al. 2014. Another white rot fungus, *Coriolus versicolor*, has shown good performance. *Coriolus versicolor* produces an extracellular laccase and extracellular enzyme. Extracellular laccase plays a role in lignin biodegradation (Aftab et al., 2011).

The optimum conditions supporting fungal growth are quite different from those favoring decolourization. The pH range for optimum growth was 4.3-4.8 and decolourization is greatly slow below pH 4 or above 5 because of poor growth. Though the fungus was grown at an optimum pH, decolourization occurs at a pH as low as 3. The decolourization is less sensitive to pH decrease than fungal growth and in the pH range of 5-7; the situation is less clear since fungal decolourization results in the formation of acids which lowers the pH rapidly (Saritha et al., 2010; Kamali and Khodaparast, 2015).

optimum temperature of the fungus growth was 40°C, whereas decolourization takes place with a little decrease in rate at temperature as low as 25°C (Tiku et al., 2010). The optimum supporting conditions of the fungal decolourization requires sufficient oxygen and a co-substrate but the addition of nitrogen source is not necessary (Verma and Madamwar, 2002; Kamali and Khodaparast, 2015).

5.2.4 Remediation with enzymes

Some of the selected enzymes like ligninase, cellulase, peroxidase etc are also showing potential to remove organic load from pulp and paper mill effluents and also having vital role to improve the quality of wastewater. Out of them the most important enzymes, especially peroxidase which is used for colour removal in bleaching effluents. It is also possible to mix enzymes together with special microbes which normally do not have high enzyme activity for decolourization process. White rot fungi uses glucose as substrate and produce peroxidase, an extracellular enzymes. It seems that this enzyme oxidizes the chromophores and removes the colour from bleaching wastewater. The colour removal from effluents at neutral pH by low levels of hydrogen peroxide (H₂O₂) was enhanced by the addition of peroxidase(Gao et al.,2010,2013;Raj *et al.*,2014).

6.CONCLUSION

In contrast of decolorization, different microorganisms show that white rot fungi particularly Phanerochaete chrysosporium, also called as model fungus and Coriolus versicolor are suitable for efficient degradation of the recalcitrant chromophoric material in bleach plant effluents. However, the requirements for high oxygen tension and a growth substrate constrain the practical implementation of fungal decolorization. Generally, Compared with physicochemical ways, biological methods for wastewater treatment are considered to be of cost benefit, eco-friendly and suitable for reduction of the BOD and COD from the effluents.

7. RECOMMENDATION

The treatment of effluents from pulp and paper mills has become an essential prerequisite prior to their discharge to receiving water bodies. In general, remedial action taken to reduce the pollution load from pulp and paper industries is of two main types:

Treatment at source process internal measures, wherein 'cleaner' technologies are to reduce the toxicity at each stage of paper making

End-of-pipe EOP treatment which deals with the effluents discharged

The Use bioremediation rather than physiochemical treatment process for waste water Treatment is more advantageous.

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