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**REVIEW OF TECHNOLOGIES USED FOR WASTEWATER MANAGEMENT IN PULP AND  
PAPER INDUSTRY**

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**ABSTRACT** The pulp and paper industry is the principal wood based paper manufacturing industry in the world. High amount of wastewater production and energy consumption are the main problems in this industry. The wastewater of the pulp and paper industry is rich in contaminants like dissolved solids, sediments, adsorbable organic halides (AOX), and chlorinated organic compounds, resulting very high values of chemical oxygen demand (COD), and biological oxygen demand (BOD). Therefore, wastewater produced in the pulp and paper making process requires treatment prior to discharge into any receiving water. This study reviews and discusses the current status of the pulp and paper industry, manufacturing techniques, key pollutants released during the production process, as well as recent and advanced wastewater treatment technologies used in the industrial sector. The review revealed that the integrated system is the most stable and appropriate system for wastewater treatment in the pulp and paper industry that can help in reducing greenhouse gas (GHG) emissions, and energy costs, and in achieving environmental regulations.

**Keywords:** wastewater treatment, pulp and paper industry, integrated system

**INTRODUCTION** The pulp and paper industry uses wood as a raw material and produces pulp, paper, paperboard and other cellulose based products. This is the principal industry in the world, producing a wood based paper product. The pulp and paper industry is one of the main industries of the North American economy and Canada ranks as the fifth largest pulp and paper producer in the world (<https://worldpapermill.com/top-pulp-paper-producing-countries>). When consider the pulp and paper industry of Canada, its annual contribution is 7,225 million CAD to real GDP, while the annual export earnings is nearly 17,500 million CAD (<https://www.statista.com/statistics/596788/key-data-on-the-paper-and-pulp-industry-in-canada>). The production increase in the pulp and paper industry to meet the human needs, after industrial revolution in particular, has created problems such as pollution of the land, water, and air. Traditionally, there are five main phases in the pulp and paper production process; raw material preparation, pulping,

bleaching, chemical recovery, and papermaking (Stenqvist, 2015). The pulp and paper mills are classified as an integrated industrial operations, which contains a process zone, utility zone and power zone (Stenqvist, 2015).

Recent studies have revealed pulp and paper industry as a serious threat to the environment due to the discharge of various toxic compound containing wastewater (Giri et al., 2014). Apart from toxic compounds, dissolved solids such as sodium chloride (NaCl), sodium sulphates (Na<sub>2</sub>SO<sub>4</sub>), calcium (Ca) and varying amounts of suspended organic materials are also very rich in pulp and paper wastewater.

Given this background, this study attempts to review wastewater treatment methods that are environmentally friendly and efficient in removing contaminants and their associated benefits. Hence, the present review focuses and discusses: (i) the current status of the pulp and paper industry; (ii) manufacturing techniques; (iii) key pollutants released during the production process; and (iv) recent and advanced wastewater treatment technologies in the pulp and paper industrial sector.

**Pulp and papermaking process** The pulp and papermaking process is divided into five major groups comprising mechanical, chemical, chemo-mechanical, thermo-mechanical pulping, and paper making (Ashrafi et al., 2015). Pulping is the initial stage of the paper making process that provides the processed material and releases a large amounts of pollutants into the environment. Also, initial stage of this process generates high amount of wastewater (Pokhrel and Viraraghavan, 2004).

**Effects of wastewater on the environment** The effluent released from the pulp and paper industry impacts all aspects of the environment such as water, air, animals and soil (Makris and Banerjee, 2002). Many researchers have reported that the presence of toxic pollutants in fish or other respiratory animals cause stress in the function of oxygenase activity and mutagenicity, liver damage, genotoxic effects, and lethal effects when the fishes exposed to pulp and paper industry wastewater (Pokhrel and Viraraghavan, 2004; Ericson and Larsson, 2000). Some of the mechanical and chemical wastewater treatment methods have created various hazards to the environment. These treatment methods mainly depend on the type and concentration of sludge retention time (SRT), nutrient availability, temperature, pH, and presence of xenobiotics (Vengosh et al., 2014). But, this is not the case for all variations; when consider other factors, reactors use for chemical treatment can be deteriorated due to extreme transient conditions that are affected by the environmental variation of wastewater treatment systems (Leitão et al., 2006). However, many of the components that are released from the secondary treatment are toxic to aquatic organisms (Makris and Banerjee, 2002).

**PULP AND PAPER WASTEWATER TREATMENT** Most of the pollution from the pulp and paper industry can be reduced by using an internal treatment process and management measures. Several studies have recommended that the changes in the internal process as a suitable measure to reduce the pollutant load in wastewater (Webb, 1994; Raghuveer and Sastry, 1990). For example, the effluent's biochemical oxygen demand (BOD) level

was reduced approximately by 60% due to a change in the internal treatment process of Irving Pulp and Paper Limited, Canada (Dube et al., 2000).

**Physicochemical treatment** These processes include removal of suspended solids, colloidal particles, floating matters, colors, and toxic compounds by using different physicochemical treatment methods. Sedimentation and floatation, coagulation and precipitation, filtration, adsorption, wet oxidation, reverse osmosis and ozonation processes are used for the treatment of pulp and paper wastewater because of their ability to remove a variety of suspended and floating materials as well as toxic compounds (Kamali and Khodaparast., 2015; Pokhrel and Viraraghavan, 2004). The sedimentation process has a possibility of removing 80% of suspended matters from wastewater and this is used as the preferred method within the paper mills in the United Kingdom (Thompson et al., 2001). Coagulation and precipitation are the tertiary treatment process in pulp and paper wastewater treatment and not usually combined with the primary treatment (Chernoberezhskii et al., 1994; Renault et al., 2009; Rohella et al., 2001). The core of this method is the addition of metal salts to the treatment tank in order to produce large flocs from small particles. In general, Wang et al. (2011) used aluminum chloride as a coagulant and a modified natural polymer as flocculants for the treatment of wastewaters from the primary sedimentation tank.

**Biological treatment** Most pulp and paper industries use aerobic, anaerobic, and integrated treatment processes to remove organic contaminants in wastewater. These methods involve the application of microorganisms such as fungi, bacteria and algae, and enzymes, as a single step treatment or in combination with other physical or chemical treatment methods. When compared with the physicochemical method, biological wastewater treatment methods are considered cost effective, environmentally friendly, and effective in minimizing the chemical oxygen demand (COD) and BOD levels in effluents (Singhal and Thakur, 2009). Both aerobic and anaerobic methods have certain advantages and disadvantages. In order to consider those benefits and drawbacks, integrated treatment (combination of aerobic and anaerobic) processes, operating under different environmental conditions, have been used to treat the pulp and paper wastewater (Pokhrel and Viraraghavan, 2004).

Aerobic process Aerobic processes are commonly used in pulp and paper mills because of their high efficiency in treating wastewater. Activated sludge and aerated lagoons are the most commonly used treatment methods among aerobic technologies in the pulp and paper industry (Mulligan, 2002; Pokhrel and Viraraghavan, 2004). The performance of the activated sludge process is varied with changes due to pH, temperature, and water and can remove 95% of soluble COD from the pulp and paper mill's wastewater (Kamali and Khodaparast, 2015). However, several studies stated that the activated sludge process effectively decreased the concentration of BOD, COD, absorbable organic halides (AOX) and chlorinated compounds in pulp and paper mill wastewaters (Table 1) (Hansen et al., 1999; Mahmood and Elliott, 2006). The aerobic process utilized *Pseudomonas putida*,

*Citrobacter sp.*, and *Enterobacter sp.* which are common microorganisms associated with the pulp and paper mill wastewater treatment process (Pokhrel and Viraraghavan, 2004). The aerated lagoons method for pulp and paper wastewater treatment was efficient in reducing the BOD by 95% and chlorinated phenolic by 85% (Pokhrel and Viraraghavan, 2004). Activated carbon and activated sludge bioreactors, a combination of biological and physical methods, efficiently remove organic substances, toxic compounds, and pathogens including adsorption onto granular carbon and biodegradation by aerobic microorganisms (Balcioglu et al., 2007). These combined treatment methods removed COD, ammonium nitrogen, and dichlorophenol by 97.2%, 99.4% and 100%, respectively, after 3 days of hydraulic retention time – HRT (Buyukkamaci and Koken, 2010; Muhamad et al., 2013).

**Table 1: Treatment performance of different aerobic methods used in pulp and paper wastewater treatment**

Treatment Method	Removal percentage		
	AOX	BOD	COD
Activated sludge	60-95	90	40-60
Aerated lagoons	15-46	50-75	30-40 (full scale lagoon) 60-70 (pilot scale lagoon)

(Adapted from Pokhrel and Viraraghavan, 2004)

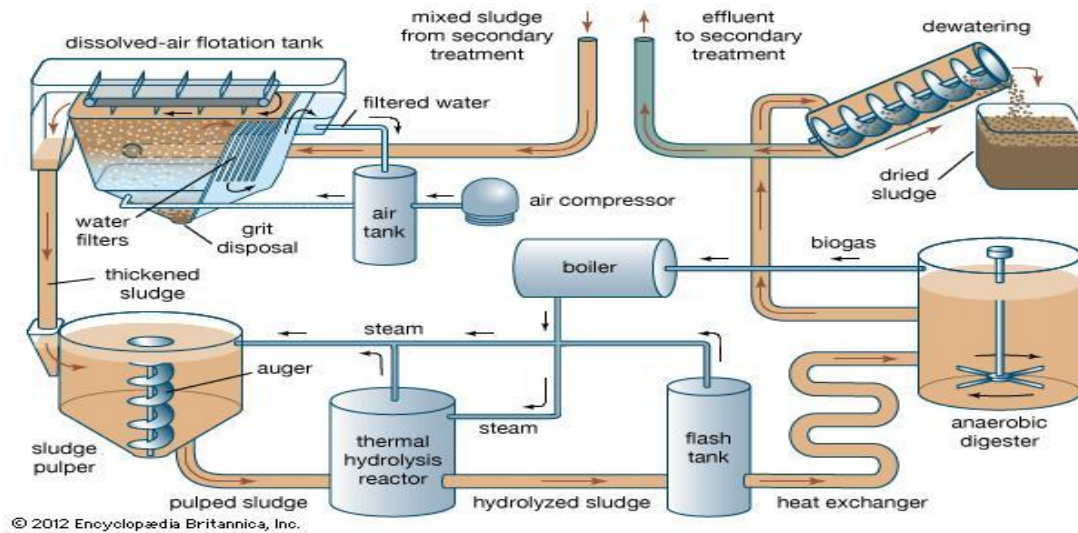
Anaerobic process Anaerobic digestion is a stable biological process that uses microorganisms, such as, *Clostridium sp.*, *Bacteroides sp.*, *E-coli*, *Acetomyces sp.* to deal with various high strength organic effluents contained in pulp and paper wastewater. Anaerobic treatment was limited in the pulp and paper industry during 1980s. However, in recent years, this treatment method has gained popularity to treat pulp and paper wastewater mainly due to its combination with conventional techniques (such as physicochemical techniques) that can reduce the sludge volume by 30% – 70% (Ashrafi et al., 2015). The management of produced sludge is one of the main problems in the pulp and paper industry; therefore, reduction of the sludge volume has direct effects on reducing the operating cost, production capital, and energy requirement.

The anaerobic filter, up-flow anaerobic sludge blanket (UASB), fluidized bed reactor (FBR), anaerobic lagoon (AL), and anaerobic contact reactors (ACR) are the most common anaerobic processes used in the treatment of pulp and paper wastewater (Ashrafi et al., 2015). Poggi-Varaldo et al. (1996) reported that continuous anaerobic treatment could remove the total COD by 48%–80% and the biodegradable COD by 87%–96% of wastewater contaminated with black liquor. Rajeshwari et al. (2000) stated that chlorine bleaching effluents were not suitable for anaerobic treatment because of their low biodegradability and presence of toxic substances that affects the anaerobic microorganisms. Moreover, high COD removal rates were obtained from

thermomechanical pulping (TMP) wastewater by using a UASB reactor at different temperature combinations such as 35°C, 55°C, and 65°C with treatment efficiencies of 82%, 92%, and 86%, respectively (Rintala and Lepisto, 1992). Kortekaas et al. (1998b) has shown that the UASB treatment is the most suitable treatment for TMP wastewater because the system is able to remove low molecular weight lignin and COD.

The yield of methane production, and minimizing of sludge from the anaerobic process is directly affected by various factors. The main influencing factors are; pulp and paper production process, reactor operating condition, wastewater composition, available inoculum for anaerobic digestion steps, and the operational cost. Even though both are considered as common greenhouse gasses; compared to carbon dioxide, methane produced during the anaerobic treatment process has a higher global warming potential. If the generated methane is used to achieve the energy requirement in the wastewater treatment process and also the other operating process of the mill, it makes the anaerobic process more efficient than the aerobic process.

Integrated process Integrated or hybrid system is a combination of aerobic and anaerobic processes (Figure 1). This system is designed to take advantage of unique features of other processes in an effort to enhance the efficiency and energy saving of the treatment process and the quality of treated effluents. The integrated system could be a combination of two physicochemical processes, or a physicochemical and a biological process, or two biological processes (Verenich et al., 2001).



**Figure 1: Integrated wastewater treatment process** (Encyclopedia, 2012)

An integrated system primarily involved in removing COD from wastewater. In addition to removing COD, other contaminants, such as BOD, AOX, color, and spores, could also be removed from the wastewater (Helble et al., 1999). However, aerobic post-treatment combined with the anaerobic UASB reactor increased the BOD removal efficiency up to 97%, compared to 83% of efficiency when using an anaerobic reactor solely (Shaw et al.,

2002). According to the findings of the Lerner et al. (2007a), the operating cost before and after the addition of UASB to the activated sludge process reduced by 50%, resulting from a lowered electricity requirement and decreased sludge production (Table 2). The combination of chemical and biological treatment methods removed 40% to 50% of COD compared with the activated sludge method (Kabdash et al., 1996).

**Table 2: Treatment performance of different integrated methods used in pulp and paper wastewater treatment**

Treatment method	Removal Percentage		
	BOC	COD	Color
<b>Combination of wet oxidation and coagulation</b>	-	51	83
<b>Combination of biofilm reactor and ozone</b>	-	80	-
<b>Combination of chemical oxidation and ozone</b>	-	50	-
<b>Combination of an aerated lagoon and a secondary clarifier</b>	96	65	22
<b>Combination of aerobic reactor and anaerobic reactor</b>	87-95	70-77	94
<b>Combination of dissolved air flotation and chemical precipitation</b>	57	92	-
<b>Combination of UASM and aerobic reactor</b>	97	72	57
<b>Combination of FBR and polyelectrolyte</b>	-	78	81
<b>Combination of anaerobic, aerobic and ozonation</b>	-	83	95

(Adapted from Ashrafi et al., 2015)

The integrated system benefits from the advantages of anaerobic processes, such as energy generation by the production of methane gas and a yielding of low sludge volume. The energy produced by burning of biogas was 2050 kWh/day (Ashrafi et al., 2015), which can be used for electricity generation for production plants or wastewater treatment plants. The cost benefit ratio of 2:3 in integrated pulp and paper wastewater treatment means that the benefit exceeds the cost by more than 50% (EU- Life - Environment Demonstration Project, 2017). The produced sludge can be used as an organic fertilizer for agriculture by using a further treatment process (Graves and Joyce, 1994). On the other hand, CO<sub>2</sub> is the main greenhouse gas (GHG) generated with a low global warming

potential in an integrated system when biogas is burnt for energy production. In addition, production and transportation of alkalinity in the anaerobic process contributes to generation of GHG, because the integrated treatment system requires a significant amount of alkalinity for anaerobic process to maintain the required pH level. Maintaining the pH level is an important factor for microorganism growth in treatment tanks. Overall, there was a  $\pm 17\%$  change in the quantity of GHG emissions in the integrated system than anaerobic treatment (Ashrafi et al., 2013b).

### Assessment of energy and electricity production in integrated wastewater treatment process

Assumptions (Al Seadi et al., 2008):

- \* Sludge volume is 5% of the total wastewater (after aerobic treatment)
- \* Wastewater discharge per day is  $10 \text{ m}^3$
- \* Hydraulic retention time for decomposing the sludge and the production of biogas is 28 days
- \* Temperature for biogas production is  $48 \text{ }^\circ\text{C}$ – $55 \text{ }^\circ\text{C}$  (Thermophilic conditions)
- \* Allocate 30% of extra volume in the anaerobic tank for sludge accumulation (70% of anaerobic digestion efficiency)
- \* If 1 kg of wastewater contains 30% of solids and it produces around 170 L of biogas
- \* Density (sludge + water) =  $1400 \text{ kg/ m}^3$
- \* Methane amount in biogas = 60%

#### Calculation:

Daily wastewater discharge =  $10 \text{ m}^3$

#### **Aerobic treatment:**

Aerobic retention time = 6 hours  
 Number of aerobic batches can be treated per day =  $24 \text{ h}/6\text{h} = 4$   
 The capacity of each set (average aerobic tank capacity) =  $10 \text{ m}^3/4 = 2.5 \text{ m}^3$   
 Daily sludge production volume =  $10 \text{ m}^3 \times 5\% = 0.5 \text{ m}^3$

#### **Anaerobic treatment:**

Anaerobic tank capacity =  $0.5 \times 28 \times 130\%$   
 =  $18.2 \text{ m}^3$   
 Daily sludge amount treated by =  $0.5 \text{ m}^3 \times 1.4 \text{ Mg}$   
 =  $0.7 \text{ Mg per day}$

The weight of  $1 \text{ m}^3$  sludge = 1400 kg  
 Solid percentage in sludge =  $(400/1000) \times 100 = 40\%$   
 Biogas production from 40% sludge =  $(170/30) \times 40 = 227 \text{ L}$   
 0.7 Mg of sludge (40% solids) = 700 kg  
 Amount of biogas production from 1 kg of sludge =  $0.227 \text{ m}^3$   
 Daily biogas production amount =  $0.227 \times 700$   
 =  $158.9 \text{ m}^3$

The amount of energy contains in 1 m<sup>3</sup> of biogas = 21.6 MJ  
 The amount of energy produces from anaerobic digestion = 21.6 X 158.9  
 = 3432.24 MJ/day  
 The amount of electricity contains in 1 m<sup>3</sup> of biogas = 2 kWh  
 The amount of electricity produces from anaerobic digestion = 2 X 158.9  
 = 317.8 kWh/day

**CONCLUSION** In this review, different wastewater treatment processes of the pulp and paper industry were evaluated and compared. Environmental performance and energy requirements play an important role in the pulp and paper industry production process. Other major concerns in the pulp and paper industry include; achieving environmental regulations, increasing the profitability, reducing the competitive market and making the pulp and paper wastewater treatment process more cost effective.

It is clear that both aerobic and anaerobic processes are more practical in tertiary treatment methods for pulp and paper wastewater treatment. However, anaerobic treatment is more powerful in the removal of lignin and (absorbable organic halides) AOX and it requires further treatment to removal of (chemical oxygen demand) COD. The application of (up-flow anaerobic sludge blanket) UASB treatment process has increased the use of different anaerobic treatment methods mainly including FBR. These processes provide significant elimination of lignin and AOX. However, biological treatment methods do not reveal an acceptable amount of color removal efficiency.

The integrated treatment system showed a greater removal efficiency of effluents than other methods. This is the best treatment methods to treat bleached kraft effluents and the end product (sludge) can be used as an organic fertilizer. However, the integrated system has the highest cost of operation for removal of bleaching kraft effluents. The greenhouse gas (GHG) production mainly depends on the type of treatment process that is used in the wastewater treatment process. There are higher GHG emissions from the anaerobic processes and integrated processes compared to the aerobic processes because of the methane production by anaerobic bioreactor. However, the produced methane can be used for energy generation which can be used to reduce the required energy consumption for both production process and wastewater treatment process. The integrated wastewater treatment process is the most stable and appropriate system for wastewater treatment in the pulp and paper industry that can reduce GHG emission, and energy costs, while achieving environmental regulations. Based on the assumptions and simple calculation, the integrated system can produce 158.9 m<sup>3</sup>/day of biogas that is equal to 3432 MJ of energy and 317.8 kWh/day of electricity with 10 m<sup>3</sup> of wastewater.

Further studies are needed to explore the cost-benefit ratio for each method used for wastewater treatment and for identifying the chemical composition of sludge produced by the integrated system. As well, further studies are needed to enhance the removal of chlorinated phenolic compound, AOX and to increase the biogas production.

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