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Treatment of Pulp and Paper Industrial Effluent Using integrated methods : A review

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ABSTRACT

The pulp and paper sector has become one of the most significant industrial sectors in the world due to its economic benefits. After primary metals and chemicals sectors, the pulp and paper industry produces the third-largest quantity of wastewater. With regard to environmental feedbacks, ongoing legal requirements, and energy efficiency measures, pulp and paper mills have recently encountered difficulties managing the ensuing pollutants. This study identify pulp and paper mill wastewaters properties, quantities and discuss the recent developments of affordable methods dealing with pulp and paper mill wastewaters. According to the results of the current study, employing integrated methods which is a mixture of treatment techniques may be more advantageous from an economic and environmental standpoint in order to reduce environmental contaminants and energy recycling

1. Introduction

The exploding world population and industrial establishment has increased which resulted in economic growth in the 21st century, causing global water pollution and water shortage problems. one of the major consumers of energy and natural resources, including water, wood, fossil fuels, and electricity, as well as a significant source of pollutants released into the environment, is the pulp and paper industries. After primary metals and chemicals sectors, the pulp and paper industry produces the third-largest quantity of wastewater.[1]. It produces 42% of global industrial wastewater as shown in (Fig. 1)[2] . It produces a lot of pollutants during the pulping and production of paper products, whose makeup varies depending on the manufacturing method[3][4], As a result, there are now significant environmental hazards due to the large wastewater discharge.[5] Recently, intensive recycling of white water is the new orientation for the pulp and paper industry to reduce fresh water uptake and meet tightened discharge standardsWhite water cannot easily be filtered due to dissolved and colloidal components , which increase electrolytes with more white water recycling, which negatively affects paper machine runability and paper quality. [6]

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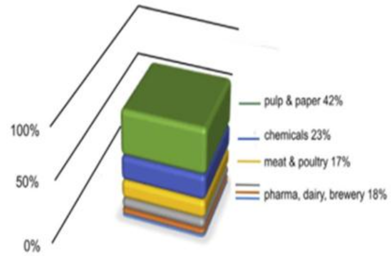

Fig. 1. Global industrial wastewater production [2]

Product demand from the paper sector has increased throughout last year. (Fig. 2). According to The Food and Agriculture Organization Corporate Statistical Database (FAOSTAT) website, In 2015, there were more than 390 million tonnes of paper produced worldwide. In addition, and according to the prognoses, paper production is set to grow. Large amounts of water are required for the pulp and paper industries. In the one ton of paper manufacturing approximately 190–200 m3 of normal water is used [7]. Frost & Sullivan forecasts that the pulp and paper industry's share of the global market for wastewater treatment will increase from $983.9 million in 2012 to $1.569 billion in 2020.

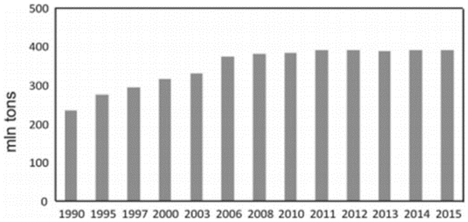

Fig. 2. Global paper and paperboard annual production in the years 1990-2015, according to FAOSTAT

1-1 The main steps in the paper manufacturing process:

Manufacturing process of paper includes the following main steps [8] :

(1) Pulping (mechanical, hybrid or chemical methods): The first step for the pulp and paper production process requires separating the cellulose fibres from the primary source, typically wood chips. Pulping processes range from the physical separation of fibers using mechanical pulping, to chemical degradation and removal of lignin to release the fibres.

(2) Bleaching: As chemical pulps are generally used for high-quality paper or personal product uses, the pulp may be bleached to enhance brightness, whiteness and perceived cleanliness.

(3) Papermaking: The stage where the paper manufacturing process begins.

1-2 Quantity of Wastewater produce from paper industries:

The pulp and paper mill industries release 75-225 m3 of effluent every tonne of paper goods manufactured (20-25 m3 from pulping and 80-100 m3 from bleaching)[9].. Several regions of the world have distinct pulp and papermaking methods. The normal amount of water used in modern pulp mills for one tonne of paper is between 10 and 50 m3.

1-3 toxic effects of paper industrial effluent:

It is important to treat industrial wastewater produced from paper and pulp manufacturing for several reasons:

1. Although the volume is relatively low, the bleaching step, particularly the alkali extraction process, often contributes the most to the overall pollution burden in the paper manufacturing process. [10]

2. Nonbiodegradable sources of colour in effluents include lignin, its derivatives, and polymerized tannins. [11]

3. Tannins absorb substantial amounts of light and heat, reducing dissolved oxygen and severely impacting aquatic plants and animals.

4. Whereas long-chain fatty acids inhibit methanogen bacteria, resin acids are toxic to fish.. [12]

5. Organochlorides bioaccumulate in aquatic organisms, especially in the body fat of tropic species that live at higher altitudes.[13]

6. The release of coloured effluents into natural water significantly alters algal and aquatic plant productivity as a result of the lower sun radiation penetration, resulting in severe aesthetic issues.

7. Resin acid, chloroform, dioxins, chlorate, chlorinated hydrocarbons, phenols, and furans are extremely hazardous to human health and can enter

the body through the lungs, causing genetic defects, cancer, and neurological conditions. [14]

8. Carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O), which are important greenhouse gases, are emitted by this business .[15]

1-4 Characteristic of waste water produced from Pulp and Paper industry:

The properties of the waste liquid to be treated depend on the type of wood, the type of operation, the amount of water the mill can cycle, the technology employed, and the managerial skills chosen.[4]. The wastewaters from high yield pulping operations contain suspended solids, organic substances, chromophoric compounds (mostly labile extractives and lignin fragments), inorganic compounds such as nitrogen and phosphorus, and salts. In addition to the foregoing, semichemical processes include lignosulfonates (produced from lignin). Solids , dissolved organics, and chromophoric chemicals are the distinguishing characteristics of Kraft process wastewaters (mainly derived from lignin). If Kraft pulps are bleached with chlorine chemicals, organochlorine compounds like dioxins and furans may be produced.[4]

The wastewater from pulping and textile production has been characterised as refractory organic wastewater with a high concentration because it has high chemical oxygen demand (COD), a low ratio of BOD/COD suspended solid (SS), salts, and colour [16]. In the textile wastewater, heavy metal ions including arsenic, lead, mercury, cadmium, nickel, cobalt, and zinc as well as colour molecules containing aromatic and azoic compounds were formed.[17]

2. Methods of treatment and its limitation

2-1 Physical methods

Physical unit operations are types of treatment where the application of physical forces is predominant. In order to remove suspended solids, colloidal particles, hazardous compounds, floating materials, and colours from wastewaters, the majority of these techniques rely on physical forces.These processes include sedimentation, screening [18],coagulation, flocculation [19], Ion exchange resin[20],ultra and nanofiltration. [21][22] [23] [24], flotation., and Electrocoagulation[25] [26] [27] [28] [29][30][9]

Table (1): summary of the chemical composition of mechanical and chemical pulping process effluents and pulp and paper mills effluents :[8][9]

For mechanical and chemical pulping process effluents		**For pulp and paper mills effluents**	
Chemical composition;	**concentration in ppm (mg/l)**	**Chemical composition;**	**concentration in ppm (mg/l)**
COD	**500 - 115000**	COD	480–4450
pH	6.3–6.8	BOD5	120-4000
Lignin	11000–25000	pH	6.1–8.3
Sulfate	3–5100	Chlorides	80–980
Sulfite	50–4800	Sulfates	241
Sulfides	1–270	Phosphates	155–470
Acetic acid	235–10400	Volatile fatty acids	950
Resin acids	3.2–550	Acetic acid	200
Chlorides	13.9–38.5	Propionic acid	98
Total acids	5	Butyric acid	36
Phenols	17–800	Total polyphenols	48
Peroxide	0–1000	Total dissolved solids	395–2500
Furfural	0–1140	Cellulose	1200
Terpenes	0.1–25000	Butyric acid	36
2-Propanol	0–18	Total polyphenols	48
Methanol	90–12000	N	Ranged from 10 to 350 according to process
Ethanol	0–3200	SS	120-4000
Abietic acid	4.3–5.2	Color (Pt-Co)	Ranged from 4000 to 5000
Oleic acid	5.3–14.5	Conductivity µs/cm	1365
β-Sitosterol	2.2	T/L mg/L Tannic acid	33
Tannins	2730		

2-1-1 Recent studies in physical methods

Table (2): summary of results of recent studies in physical treatment methods:

Treatment process	**wastewater Source**	**removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
Granular ion exchange resin	Paper mill				DOC: 72 %	[20]
activated carbon					DOC: 76%	
nanofiltration					DOC: 91 %	
Ultrafiltration.	Pulp & paper	89%		95%	Hardness: 83% Sulfate: 97% Conductivity: 50%	[21]
Membrane filtration	Pulp & paper				membrane flux performance study.	[31]
Nanofiltration, with pH adjustment	Paper mill	65-98%		90-98%	TSS: 66-100%	[22]
Colloid-enhanced ultrafiltration with surfactant-polymer complexes	Pulp & paper				Cl-phenols: 90-99%	[23]
nano- and reverse osmosis filtration for of biologically treated effluents	Pulp & paper	66-100%	53-94%	97-100%	Monovalent: 95%	[24]
Electrocoagulation	Paper mill	55-75%	70-80%		Lignin:80-92% Phenol: 93-98%	[26]
Electrocoagulation	pulp and paper industry wastewater	**77%**		**99.6%**	**TOC 78.8%**	[27]
Electrocoagulation	pulp and paper industry wastewater	68%		94%		[32]
Electrocoagulation	Pulp and Paper Industry	**84%- 85 %**				[30]
Electrocoagulation	Wastewater from recycling paper	**79.5%,**		**98.5**	**TSS 83.4%,** **ammonia 85.3%,** **TDS 35% turbidity 99%,**	[29]
Electrocoagulation	effluent from the pulp and paper industry	**82%**		**>99%**		[33]
Electrocoagulation	Kraft paper mill	**BI increased to 0.41 BOD5/COD**		**>95%**	**tannin/lignin were >70%**	[9]
Electrocoagulation	Pulp & paper				Sulfides: 88% Phosphorus: 40%	[34]
Electrocoagulation	Pulp & paper	77- 80 %		90 -91 %		[14]
granular activated carbon after Al and Fe electrodes electrocoagulation	Cardboard	75% - 79%	99.9%			[30]
Electrocoagulation (Fe)-flotation Followed by photocatalysis with UV and TiO2	Pulp & paper	COD: 88% BOD/COD: 0.15-0.89				[35]
Electrochemical oxidation	Lignosulfo-nate				TOC: 80%	[36]
Electrochemical treatment (anodic oxidation)	Paper mill	97%		53-100%		[18]
Electrocoagulation	Pulp & paper	77%-80 %		90-91%		[37][38]
Electrocoagulation with Al and Fe electrodes	Paper mill	55-75%	70-80%		Lignin:80-92% Phenol: 93-98%	[26]
Electrocoagulation	Pulp mill				Toxicity: 100% Resins: 63-97% Copper: 80-100%	[34]

Electrocoagulation with aluminum or iron plates	Paper mill	32-68%			DOC: 24-46%	[39]

2-1-2 Advantages and disadvantages of physical treatment methods:

2-1-2-1 Electrocoagulation

Electrocoagulation is Simple methods that is , efficient, affordable, simple to use, and environmentally friendly .Additionally, filtered water produces less sludge and is potable, clean, colourless, and odourless.These methods completely exclude the possibility of secondary water contamination [40][41].

On the other hand Electrocoagulation have a main disadvantage such as using High current densities (5–35 mA/cm^2) in these tests, which resulted in energy loss, the heating of waste streams, and a decline in operational efficiency.

The performance of EC at low current densities (5 mA/cm2) has not been studied, despite Mollah's observation that electrodes with a greater surface area are needed to produce a feasible rate of metal dissolution at low current density.. Due to the huge energy input necessary—which can potentially exceed 96 kW h/kg of COD removed—this results in a very high cost of treatment. [42]

2-1-2-2 filtration

compared to water evaporation or centrifugation, likely to be less energy-intensive.

Some P&P plants have adopted membrane systems to comply with regulatory requirements governing wastewater discharge.in addition to take the necessary steps such paying attention to supplying a consistent supply of acceptable sweetener fibre, attention to providing separate impurities smaller than approximately 100 m in diameter, and use of an efficient retention aid system, solids losses at that point in the paper machine system can be reduced [43][44]

On the other hand filtration have a main defects on membrane technology for the treatment of various forms of wastewater has been fouling (The clogging or blocking of pores). [45]. Flux drop as a result of membrane fouling is a serious restriction associated with the use of membrane technology. Despite numerous attempts, this issue is still unresolved andhas various operational ramifications as well as cost-prohibitive maintenance, especially at paper mills where wastewater calcium concentrations are typically high.

2-1-2-3 Flotation

Flotation is simplicity, flexibility, low energy use, small space requirements, low sludge volume, operating selectively, and high efficiency methods that are desirable. The recovery of valuable metals, the complete separation of many ions, the pre-concentration of rare earth elements, wastewater treatment, etc. have all benefited from the application of ion flotation.but high initial expenses , energy costs ,costs of operation and maintenance that are not minimal are considered the worst shortcomings for floatation in addition to . Chemicals that are Selectivity depends on pH[46]

2-2 Chemical treatment methods (ADVANCED OXIDATION SYSTEMS)

Chemical unit processes, including as precipitation, gas transfer, adsorption, and disinfection, are treatment techniques that remove or convert contaminants by adding chemicals or by causing another chemical reaction.

Oxidation is the process of transferring electrons from a reductant, which is an electron donor, to an oxidant, which is an electron acceptor and has a higher affinity for electrons. Both the oxidant and the reductant undergo chemical alteration as a result of these electron transfers, sometimes creating chemical entities that have an odd number of valence electrons. Since one of these species, often referred to as radicals, has an unpaired electron, it tends to be highly unstable and thus highly reactive. reactions of oxidation that further oxidation events between the radical oxidants and tend to occur after the production of radicals.organic and inorganic reactants until thermodynamically stable oxidation products were formed.. [4]

AOPs include ozone [47][48][49][50] [51]

,Wet oxidation [52][53] [54]

different oxidising species combined with catalysts or UV light [55][56][57][58][59][59] [60], hydrogen peroxide [61], and The Fenton technique, which uses metal ions (Fe_{2+}) as homogeneous catalysts to transfer electrons from H2O2 to catalyse catalytic oxidation [62][63][64][65] [66][67][67]

2-2-1 Recent studies of chemical treatment methods

Table (3): summary of results of recent studies in chemical treatment methods:

Treatment process	**wastewater Source**	**removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
1wt% $ZnAl_2O_4/BiPO_4$ under UV light (UV/ZB_3) **(photo-Fenton processes)**	textile wastewater	CODCr =64.34%		58.28%		[68]
	eucalyptus chemimechanical pulp wastewater (ECMPW)	CODCr =59.23%		52.47%		
Wet oxidation	Paper bleaching effluent				AOX and Cl– removal yields were 83–90 and 73–76%, respectively, for all types of catalysts	[52]
Dual flocculation of pomegranate seeds and alum	pulp and paper wastewater	81%			Turbidity 98%	[69]
photo-Fenton treatment	board paper mill	80%				[70]
UV		11				
UV/ H_2O_2		33%				
UV	effluent from a board paper mill	increases of the BOD5/COD by 0.25				[70]
UV/H_2O_2		increases of the BOD5/COD by 0.45				
photo Fenton processes		increases of the BOD5/COD by 0.70 cod removal (76%)				
Fenton method	pulp and paper wastewater	75%		98%	aromatic compound removals 95%	[71]
wet oxidation using catalyst (5% CuO/95% activated carbon)	pre-treated P&P mill	89%				[53]
Alum & cationic PAM flocculant	Pulp & paper				TSS: 99% Sludge vol: 37 mL/g	[19]
Alum & cationic PAM flocculant; PAC & anionic PAM flocculant	Pulp & paper	91-91%			Turbidity: 99.8-99.9% TSS: 99.4-99.5%	[72]
Ozone or UV exposure of mill water & model compounds	Cardboard	18-99%	37-65%			[47]
Poly-DADMAC & polyacrylamide	Pulp & paper	68-98%			Turbidity: 50-95% TSS: 60-94%	[73]
pretreatments for algal treatment using Oxidation and catalytic oxidation	CEH bleach	86-90%		96-99%		[74]
Ozone	Paper recycle mill	51%				[48]
aluminum chloride Coagulation and adsorption on tuff, followed by nanofiltration membrane	Wood & pulp				Total carbon: 67% TOC: 77% Inorganic C: 49%	[75]
Ozone; Ozone & biological; Biological, ozline, & biological; NF & ozline & biological	Kraft ECF	17-65%			TOC: 5-50% Color: 80%	[49]
UV/TiO2 & UV/ZnO on Al foil or luffa followed by biological	Bleaching				TOC: 96% Cl-phenols: 90-99%	[56]
utilising a Pd/AC reactor for catalytic hydrogenation in a trickling bed reactor.	ECF bleach	12%	Went up 47%	61%	Ecotoxicity: 70-98% AOX: 85%E1, 23%D	[63]
Advanced oxidation with UV, UV-H_2O_2, and UV-TiO_2	Pulp mill				TOC: 80% Toxicity: 94%	[57]
Ozonation with catalysts (Fe-Mn/ sepiolite)	Pulp & paper	58%			pCP: 98%	[50]
UV photodegradation with or without TiO_2 catalyst	Pulp mill				Lignin: 30-70%	[58]

Sulfuric acid treatment (pH 1 or 3) proceeded by ozonation at pH 1-12	Pulp & paper	77%		96%		[76]
Wet oxidation (with or without CuO/CeO_2 catalyst), proceeded by coagulation with $FeCl_3$ or PAC	Pulp mill	51-77%		71-87%		[54]
Thermochemical precipitation with $CuSO_4$ catalyst and others	Pulp & paper	63%		92%		[77]
Catalytic wet oxidation under moderate conditions	Pulp & paper	61-89%				[53]
Solar catalytic treatment with TiO_2	Paper mill	75%			TSS: 80%	[55]
TiO_2 / solar UV	Bio-treated effluent	83%				[60]
Conventional Fenton and UV Fenton oxi retentate from Reverse osmosis system	Newsprint paper mill	80 to 100%				[65]
Coagulation and flocculation	Newsprint paper mill	50%			SiO2 removal: up to 100%	[65]
Photocatalytic treatment with UV and TiO_2 or ZnO	Pulp & paper	66-90%	78-84%			[78]
Photocatalytic treatment with UV & TIO_2 and optional peroxide	Paper mill	54-65%		82-89%		[79]
Solar Fenton treatment and dark Fenton	Pulp mill	90%			Polyphenols: 90%	[66]
Advanced oxidation with ozone or TiO_2 and irradiation	Pulp & paper	COD: 35-60% COD: 90% as post				[80]
Heterogeneous photo-Fenton and photocatalysis	Lignin	20-80%				[67]
TiO_2/Fe(III)/solar UV oxidation	Pulp mill			78%	TOC: 64% AOX: 68%	[81]
Coagulation with poly-DADMAC of various molecular mass	Pulp & paper	90-99%			Turbidity: 70-92%	[73]
Water coagulation following bio-treatment with chitosan and PAC	Recyc. paper	40-80%			Turbidity: 55-85%	[82][83]
Coagulation-flocculation with chitosan and $FeCl_3$	Pulp & paper			90%	Turbidity: 89% Lignins: 70-80%	[35]
Coagulation-flocculation, using $FeCl_3$ & chitosan, proceeded by UV/TiO_2/ H_2O_2 with mercury lamps	Pulp & paper	BOD/COD: 0.71		98-100%		[35]
Coagulation with alum or PAC and a flocculant	Pulp & paper	60%			Turbidity: 98% TSS: 92%	[84]
UV oxidation with TiO_2 & H_2O_2	Bleachery				AOX: 80-90% Lignin: 22-88%	[59]
Coagulation of aluminium chloride using a flocculant based on starch	Pulp mill				Turbidity: 99.6% Lignins: 88%	[85]
Flocculation using polyacrylamide of various charge	Pulp & paper	93%			Turbidity: 95% TSS: 98%	[19]
Photocatalytic treatment with TiO_2 on activated carbon support	Paper mill	62%				[86]
Alum-based coagulation and polyacrylamide-based flocculation	Paper mill	26-97%	26-87%		TSS: 76-99% NH3: 6-57%	[87]

2-2-2 Advantages and disadvantages of chemical treatment methods:

2-2-2-1 Ozonation

a strong oxidant, ozone (O_3) is always produced on-site and is very effective in killing bacteria, viruses, and protozoa. Transporting or keeping risky materials on hand is not necessary.Compared to chlorine disinfection, ozone produces fewer THM disinfection byproducts (but bromate may be formed). Waste water that have undergone this disinfection process don't contain chlorine or other chlorinated disinfection byproducts, therefore dechlorination is not essential. On the other hand undesired secondary reactions of ozone with the by-products lead to the formation of new hazardous chemicals such ketones, organic acids, and aldehydes[88]. Ozone solubility is constant at a given temperature, therefore when the liquid is saturated, an excess dose of ozone does not worsen the discolouration. The amount of HO• radicals and ozone in solution are essentially constant when ozone concentration is higher than the optimal dose[4].

2-2-2-2 UV-catalysis

The effectiveness of ultraviolet radiation alone is typically lower than that of other AOPs, however it can be significantly boosted when combined with other AOPs, such as UV-ozone, H_2O_2, or specific catalysts (metal salts and semiconductors)[43]**.** But Merayo and Hermosilla observed that ozone and photo-Fenton treatment were extremely successful as a post-treatment more than UV-catalysis[51] [65]

2-2-2-3 Hydrogen Peroxide

hydrogen peroxide doesn't leave behind any stains or fumes. Since hydrogen peroxide is entirely soluble in water, safety relies on the applied concentration but The efficiency of the overall deterioration is impacted by this reaction, which also serves as a radical scavenger, raising operating costs significantly. [89], and deterrent

effects on compound degradation . Since hydrogen peroxide alone is unable to oxidise the dissolved substance, radicals should be created through the direct reaction of another oxidant [like ozone] with the ionic form of hydrogen peroxide

2-2-2-4 Fenton methods

The reagents are harmless to the environment and safe to handle. It is technologically feasible for direct use on any scale because the oxidation process doesn't require very complex devices or pressurised systems (laboratory industry)[90] Additionally, it can be employed as a pre-treatment for the biological stage to improve the biodegradability of recalcitrant substances and lessen the toxicity of the effluent. [91] and for the removal of the polyphenols and organochlorine chemicals found in the effluents from several bleaching operations [57]. One the other hand - Hubbe noticed that although while Fenton processes appear to work well in laboratory settings, they are not frequently used in industrial settings because ozone methods have far more experience being put into use [92]. The growing availability of ozone treatments for bleaching stages in paper mills as well as the difficulties of using Fenton processes, such as the production of iron sludge and the requirement for acidic conditions for optimal operation, are other causes. These problems can be avoided by applying these treatments at neutral pH or by utilising heterogeneous catalysts. [43] [67][65]**.**

2-2-2-5 Wet oxidation with O2

catalysts are promising materials, because they are able to promote the oxidation in a shorter time under mild reaction conditions[93] but reaction conditions make the method economically unfeasible and it is necessary to lower the reaction temperature.

2-3 Bioogical treatment methods

Biological unit processes are treatment strategies that involve biological activity to remove pollutants. [11]- biological processes are divided into five categories: aerobic, anoxic, anaerobic, combined aerobic, anoxic, anaerobic processes, and pond processes.

2-3-1 Aerobic biological treatment methods

In P&P mills, aerobic biological wastewater treatment is commonly used. . Reducing the BOD levels of the treated wastewater is one of the main goals of aerated biological treatment. For the removal of colour, which would necessitate the breakdown of recalcitrant compounds, such treatments are typically less effective.Aerobic methods include Activated [94][95] [96],Aerated Stabilization Basins [97],SBR[98],Membrane bioreactor (MBR) [99][96],packed bed reactors [100],Aeration pond.

2-3-1-1 Recent studies of aerobic biological treatment methods

Table (4): summary of results of recent studies in aerobic biological treatment methods:

Treatment process	**Source of wastewater**	**Contaminants removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
Activated sludge	Paper mill whitewater	74-95	——	76	——	[94]
Activated sludge	Kraft pulp mill	60	90	40	36 (Tannin and Lignin)	[101]
Activated sludge	Integrated pulp mill	60-70	95	——	60 (TOC)	[95]
Activated sludge	pulp-and-paper wastewaters	70%	90%		60% AOX removals	[96]
Multiple stage (AS)	Black liquor	65	95	——	——	[102]
Aerated Stabilization Basins	pulp-and-paper wastewaters	30%-40%	50%-70%			[103][97]
Aerated Stabilization Basins	pulp-and-paper wastewater	up to 67%				[103]
AS, facultative stabilization basin (FSB) and ASB	Kraft mill wastewaters	up to 70%			56% AOX	[104]
SBR	CTMP and TMP	53-62	88-94	——	——	[98]
SBR	Hardwood Kraft mill	69	——	——	> 80 (TSS)	[105]
SBR	(CTMP) wastewater		77%			[98]
Membrane bioreactor (MBR)	Paper mill	80	97	——	> 90 (TSS)	[96]
Membrane bioreactor (MBR)	Paper mill	92	> 98	——	84 (Ammonia), >99 (TSS)	[99]
Facultative stabilization basin (FSB)	Kraft mill	62	> 95	——	51 (AOX), 69 (chlorinated compounds)	[104]
activated carbon sequencing batch biofilm reactor (GAC-SBBR)	recycled paper wastewater	94.8			100 % NH_3–N and 80.9% 2,4-DCP removal percentages.	[106]
GAC-SBBR	P&P mill wastewater	92 ±6			2-CP 99±1 2,4-DCP 68±1 2,3,4,5-Te CP 4276 mg/L 97±6	[107]
GAC-SBBR	Recycled paper industry	(80.1 %) at COD:N:P ratio of 100:5:1				[108]
packed bed reactors	olive pulp				12.65 g phenol/(l d)	[100]
NaBH4 was used as a reducing agent in a pre-treatment to simulate a continuous stirring batch reactor.		95%	98%		97.5% color reduction 97% TSS reduction	[64]
Aeration pond	P&P mill wastewater				2,4-DCPat initial concentration 20 µM is 77.6 % 2,4-DCPat initial concentration 160 µM is 77.6 % 2,4-DCPat initial concentration 200 µM is 77.6 %	[109]
Aerobic biological treatment	Pulp mill	65-71%			AOX: 38-43%	[110]
Mixed culture of three bacterial strains	Pulp mill	91%	93%	96%		[111]
Activated sludge treatment for 280 days.	Pulp mill	60%	95%			[101]
Fungal treatments (white rot and soft rot)	Bleaching	74-81%		72-74%	Lignin: 25-46%	[112]
Bacterial treatment	Pulp & paper			27-30%	Dechlorination: 59%	[113]
Peroxidases form potato pulp	Phenolic				Phenols: 90-95%	[114]
Activated sludge treatment	Pulp & paper				Most of the contents were of medium molecular mass	[95]
Aerated biological treatment	Pulp mill				Phyto-sterols: 90%	[106]

Fungi isolated from polluted soil were used as an immobilized fungal consortium.	Pulp & paper	89%		79%	Lignin: 79%	[115]
Aerated granulated carbon sequencing batch biofilm reactor	Recyc. paper	34-90%			Cl-phenols: 90% Nitrogen (NH3): 90%	[108]
Pilot-scale aerobic lagoon treatment	Paper mill		73-93%		Color: Increased	[116]
Granulated activated carbon sequencing batch biofilm reactor	Recyc. paper	53-92%			AOX: 26-99%	[107]
White-rot fungal treatment and Surfactants	Pulp & paper	41% -75%		34% - 81%	Lignin: 16%- 66%	[117]
Activated sludge & options with stabilization basins	Bleachery				Cl-phenolics: 85-93% AOX: 43-58% Toxicity: Removed	[104]
Four fungi were compared	Pulp & paper			67%	Lignin: 37% Toxicity: 60%	[118]
Algae batch reactor	Pulp & paper	84%			AOX: 80%	[74]
Black liquor treatment with white-rot fungus on porous plastic support with biofilm.	Pulp mill	48%			Lignin: 71%	[119]

2-3-1-2 Advantages and disadvantages of aerobic biological treatment methods:

2-3-1-2-1 Activated sludge

Installing the Activated sludge system doesn't cost much, and the initial investment gets good value.

As long as the sewage is of a consistent kind and volume and the activated sludge remains activated, high-quality effluent water is produced. But unfortunatly Activated sludge is sensitivity to shock loading and toxicity, as well as the limited ability to remove non-biodegradable materials,. [29]. nutrients must be supplied as a preconditioning step before biological therapy to meet the needs of bacterial growth.[17]

2-3-1-2-2 Sequencing Batch Reactor (SBR)

Sequencing Batch Reactor is a Good settling reactor , no sludge storage, Typically for smaller plants

And frequently no primary clarifier but Special aeration equipment and Need to recycle early are main disadvantages

2-3-1-2-3 Membrane bioreactor (MBR)

Membrane bioreactor reduce the need for settling basins or clarifiers and their associated space and financial costs, or making it possible to handle wastewater in a smaller area. [120]. Other advantage like High effluent quality. No secondary clarifier. and Maintains high MLVSS is provided in Membrane bioreactor .

On the contrary , high startup costs ,suction on a tiny filter requires additional power and cleaning a membrane process that takes time For a membrane system are the main disadvantage of Membrane bioreactor

2-3-1-2-4 packed bed reactor

packed bed reactors are Low costs for building, running, and maintenance. effective at high temperatures and pressures. Completes sludge treatment and is ideal for small settlements and tropical regions. On the other hand, Temperature is difficult to control. Catalyst replacement is challenging. Gas stream channelling can happen, creating inefficient areas in the reactor are the main disadvantages.

2-3-1-2-5 Aeration pond

Aeration pond is low cost methods that are low maintenance, easy to build Completes sludge treatment and is ideal for small settlements and tropical regions. But Aeration pond Requires a large land area and not recommended For large discharge are the main disadvantages.

2-3-2 Anaerobic biological treatment methods

Anaerobic baffled reactors [121], anaerobic filters ,upflow anaerobic sludge bed reactors,[122]

[123] and anaerobic membrane bioreactors are the basic anaerobic reactors used to treat P&P mill wastewaters to date [43] . Reduced hydraulic retention time allows for faster treatment of larger volumes of wastewater in a given length of time, which was the fundamental motivation behind the invention of such high-rate reactors

2-3-2-1 Recent studies of anaerobic biological treatment methods

Table (5): summary of results of recent studies in anaerobic biological treatment methods:

treatment process	**wastewater Source**	**removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
Upflow anaerobic sludge blanket UASB	Kraft pulp mill	79			71 – 99.7 (chlorinated compounds)	[122]
UASB	Bagasse wash	80 – 85				[124]
UASB	bleached and unbleached Kraft mill wastewaters	79% to 82%			71% to 99% chlorinated organics	[122]
Upflow anaerobic sludge blanket UASB	Kraft pulp mill	79			71 – 99.7 (chlorinated compounds)	[122]
Up-flow anaerobic filter	Bleaching process	50	% 70		67 (AOX), 86 (Lignin), 63 (Phenol)	[123]
a modified anaerobic baffled reactor	recovered fibers mill wastewater	71%	71%		50% TDS removal ratio 45% TSS removal ratio 49% TS removal ratio 45% VSS removal ratio the daily methane ranged from 0.003 to 0.09 L CH_4/g COD	[121]
Thermophilic submerged aerobic membrane bioreactor.	P&P mill wastewater	92-93%				[125]
Mesophilic & thermophilic filters were compared.	Paper mill	80%				[126]
Anaerobic biological treatment alone	Kraft pulp	61%	90%		TOC: 69% AOX: 55%	[127]
Anaerobic digestion of wastewater or sludge	Pulp & paper	30-90%			Vol. solids: 21%-55%	[128]
Anaerobic co-digestion with MSG and sludge from pulp & paper	Pulp & paper	48%				[129]
Anaerobic digestion of TCF bleaching effluent	Pulp & paper				Biogas yield was optimized.	[130]
Anaerobic baffled reactor	Recyc. Paper	85%				[121]
Upflow anaerobic filter treatment of bleachery effluent	Pulp & paper				AOX: 28-88% Enhanced to 90-93%	[123]
Thermophilic submerged aerobic membrane bioreactor.	P&P mill wastewater	92-93%				[125]

2-3-2-2 Advantages and disadvantages of anaerobic biological treatment methods:

anaerobic biological treatment methods have many advantages like fewer greenhouse gas emissions (250 kg CH_4/day in anaerobic and hybrid systems) when compared to aerobic processes, as well as the creation of CH_4 or H_2 as an energy carrier. [2]. At rates of 70–90% and 90%, respectively, anaerobic digestion has been employed to remove COD and adsorbable organic halides. [120] . Sludge production is often lower than in traditional aerated biological treatment systems. the ability to reduce the volume of produced sludge by 30–70%, low capital and operating costs, the ability to be applied at different scales, and the rate of pathogen destruction

on the other hand anaerobic biological treatment methods have many disadvantages like The possibility of sulphate ions being reduced to other substances, such as sulphides or H_2S, is an issue that is inherent to anaerobic processes. . These substances are produced as a result of sulfate-reducing bacteria. Reduced sulphur compounds may prevent methane from being produced. Biogas combustion is a necessary additional step for the conversion of biogas to energy (the conversion efficiency of methane to electricity ranges from 30% to 40%). The anaerobic digesting processes have lengthy start-up times (months)

2-4 Integrated treatment methods

There is an ideal combination of circumstances for every treatment, which can then be arranged in many ways. As a result, adjustments to the parameters at each stage can quickly address changes in the effluent's input composition, giving the process a wide variety of options and a flexible design.

The use of integrated systems, often referred to as hybrid systems, has drawn a lot of interest in an effort to boost treatment effectiveness and raise effluent quality. Two physicochemical processes, a physicochemical and a biological process, or two biological processes could all be combined to create the integrated system.

2-4-1 Recent studies of Integrated treatment methods

2-4-1-1 Chemical methods integer with physical methods

Problems with the traditional Fenton process, which uses a homogeneous catalyst, include a slower rate of ferrous ion regeneration and an excessive amount of iron sludge creation. But these issues must be resolved for this method to be commercialize

Table (6): summary of results of recent studies in Integrated treatment methods (Chemical methods integer with physical methods)

Treatment process	**Source of wastewater**	**Contaminants removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
Composite flocculant preceded by reverse osmosis filtration	Pulp & paper	75%			The composite flocculant aided the reverse osmosis process.	[131]
Membrane filtration and ozone treatment of the concentrate	Pulp & paper	83-97%		96-99%	Turbidity: 50% Lignin: 50% Sulfate: 88-98% Salts: 76-92%	[132]
Microfiltration & electrodialysis with ion exchange pairs	Pulp & paper				Salts: 95% Lignin: 90%	[133]
Nanofiltration and ultrafiltration using shear-enhanced modules	Pulp & paper			86-98%	Salts: 1-78% TOC: 25-88% Sugar: 36-97% Lignin: 17-97%	[134]
Ultrafiltration with polymer complexation by PEI, PVOH	Pulp & paper	45-57%		88-98%	Metals: 35-92% Turbidity: 99%	[135]

2-4-1-2 Chemical methods integer with biological methods

Integrating chemical oxidation and biological treatment would be more practical given their respective benefits and drawbacks. In the combined process, the persistent organic pollutants would first undergo chemical oxidation to become more biodegradable intermediates, which could then be easily eliminated in a following biological stage.

Table (7): summary of results of recent studies in Integrated treatment methods (Chemical methods integer with biological methods)

Treatment process	**Source of wastewater**	**removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
After activated sludge, combined ozone and fixed bed biological post-treatment of effluents	Paper mill	88			By adjusting the ozone dosage, the BOD/COD ratio may be optimally achieved.	[136]
Integrated ozonation with bio treatment; focus on recalcitrant organic matter	Pulp mill				High MW degradation increased from 5% to 50% 30% greater TOC mineralization.	[137]
Yeast isolates combined with solar and dark Fenton	Pulp mill	68%			Polyphenols: 27% TOC: 90% (Fenton)	[138]
The chromophores were reduced using sodium borohydride, preceded by aerobic bio-treatment	Pulp & paper	35-92%	to 99%	97%	TSS: to 97%	[64]
Integrated photo-catalytic (TiO2) and biological treatment	Pulp mill				The photo-oxidation decreased the time (64 %)of bio treatment.	[139]
Comparing advanced oxidation systems for subsequent biodegradability	Paper mill	80%			TSS: 97%	[70]
Ozone & biofilter	Paper industry	60-85%			O3+bio-filter+O3+bio-filter improved COD removal >10% reducing the need of ozone.	[140]
Thermophilic submerged aerobic membrane bioreactor	Pulp & paper	87%-96%		Up to 100%		[125]
Advanced oxidation with H2O2/UV as post-treatment after anaerobic treatment	Kraft pulp	0%-11%			Lignin: 16-35% AOX: 23-54%	[127]
Electrochemical pretreatment to improve biodegradability	Pulp & paper	55%		87%		[141]
Horseradish peroxidase and H2O2 treatment of foul condensate	Kraft pulp	marginal			Phenols: 90-100% Toxicity: 40-50%	[142]

2-4-1-3 physical methods integer with biological methods

Table (8): summary of results of recent studies in Integrated treatment methods (physical methods integer with biological methods)

Treatment process	**wastewater Source**	**removal efficiency (%)**				**References**
		COD	**BOD**	**Color**	**Other compounds**	
Electrocoagulation & electro-oxidation, then bio treatment	Bleaching				Cost: 41% vs. usual coagulation/biological	[143]
Biotreatment, followed by nanofiltration	Pulp & paper	91%			Hardness: 92% Sulfate: 98%	[144]
Laccase polymerization was followed by membrane filtration	Pulp & paper	60%				[145]
Anaerobic, aerobic, ultrafiltration, reverse osmosis sequence.	Recyc. paper	53-81%	68-98%		Sulfate: 30-96%	[146]
Anaerobic/aerobic membrane bioreactor with NF or RO filtration	Paper mill	96-98%				[147]

3. Summary and future research needs

According to the current review and previous studies, integrated methods for pulp and paper wastewater treatment have become more widespread. integered methods have achieved more efficiency and cost reduction. More advanced studies should be studied, especially in combination with new chemical, physical, and biological methods, and develop the current methods to increase the removal of pollutants, especially COD, turbidity, and lignin, from the P&P mill wastewater effluents. Scientists candidate physical-chemical methods(sedimentation-filtration methods) as the future methods for adapting factories operating conditions because of their low cost. however, require further research. especially for coagulant type in sedimentation process and filtr media in filtration process should be studied to increase removal of pollutants especially, COD, turbidity, and lignin from the P&P mill wastewater effluents.

4. Conclusion

The purpose of this study was to evaluate the existing state of wastewater treatment for pulp and paper in order to help the P&P industry choose and implement an appropriate approach. The pulp and paper sector is currently undergoing significant reforms in both environmental performances and production methods in order to comply with strict environmental requirements, preserve their profitability, and exceed decreasing and competitive markets. There are a number of physicochemical and biological techniques (integrated methods) that have been widely used to treat wastewater from pulp and paper mills more effectively and economically. Various suspended and floating substances, as well as refractory contaminants, have been removed from produced wastewaters using physicochemical techniques. P&P mills have employed sedimentation widely. The effectiveness and necessity of pre-sedimentation when it has been carried out before filtration.

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