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Additive and additive-free treatment technologies for pulp and paper mill effluents: Advances, challenges and opportunities

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ABSTRACT

In the present manuscript, novel effluent treatment processes for pulp and paper mill effluents are divided into two categories: a) those involving the use of chemical additives and b) those which are free of such chemicals. It is especially of high importance for pulp and paper industry to adopt the most efficient and cost-effective treatment methods. This paper critically reviews the recent studies on the treatment of pulp and paper mill effluents while providing suggestions for further studies on the application of various physic-chemical and biological methods for the treatment of such complex effluents containing a number of recalcitrant pollutants.

1. Introduction

It is well known that the effluents from pulp and paper mills (PPMs) are highly polluted by various types of recalcitrant organic pollutants such as adsorbable organic halides (AOXs), as well as inorganic chemicals normally added during the process of producing pulp and/or paper [1–3]. However, the amount of pollutants loaded to the final effluents is highly dependent on various parameters such as the type and the stage of the manufacturing process and the type of the raw materials used for the production of pulp and/or paper [4]. Due to this fact, there is a need for treatment methods which are efficient enough, while being both cost-effective and environmentally friendly. It is also of high importance to emphasize that the effluents treatment technology to be adopted by the industry must have the ability to be easily transferred from lab-scale installations to full-scale applications. This is one of the most important issues that must be taken into consideration because a number of methods developed so far for the removal/degradation of recalcitrant compounds were used in lab-scale experiments but with a limited number of evidences for a rapid transferring of the newly developed techniques to real-scale applications [5]. There are a number of barriers for the commercialization of the novel wastewater treatment technologies. The expertise and financial supports required (for instance in terms of the initial investments), the insufficiency of the regulations especially in the developing countries which are not able to force complying with the scientific-based environmental standards, and the treatment costs associated with the application of advanced treatment methods are among

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Nomenclature

AD	Anaerobic Digestion
AO	Anodic Oxidation
AOPs	Advanced Oxidation Processes
AOX	Absorbable Organic Halides
AS	Activated Sludge
BDDs	Boron-Doped Diamond electrodes
BMP	Biochemical Methane Potential
BOD	Biological Oxygen Demand
COD	Chemical Oxygen Demand
CTMP	Chemical Thermo-Mechanical Pulp
CWAO	Catalytic Wet Air Oxidation
CWPO	Catalytic Wet Peroxide Oxidation
DC	Dc power
DCS	Dissolved and Colloidal Substances
DSA	Dimensionally Stable Anodes
ECF	Elemental Chlorine Free
EFB	Empty Fruit Bunch
EAOPs	Electrochemical Advanced Oxidation Processes
FTOC	Filtered Total Organic Carbon
GAC	Granular Activated Carbon
GAC-SBBR	Granular Activated Carbon Sequencing Batch Biofilm Reactor
HRT	Hydraulic Retention Time
HW	Hardwood
MBT	Multi-Barrier Treatment
MTBE	Methyl-Tert-Butyl Ether
NF	Nanofiltration
NSSC	Neutral Sulfite Semi-Chemical
OLR	Organic Loading Rate
PAC	Polymeric Aluminum Chloride
PAM	Polyacrylamide
PCP	Pentachlorophenol
PHA	Polyhydroxyalkanoate
PMAC	Poly methacryloxyethyl-trimethyl ammonium chloride
POME	Palm Oil Mill Effluent
PPMs	Pulp and Paper Mills
PPMW	Pulp and Paper Mill Wastewater
P&P	Pulp and Paper
PS	Poly-Silicic acid
RFP	Recycled Fiber Pulp
RO	Reverse Osmosis
SCOD	Soluble Chemical Oxygen Demand
SGBR	Static Granular Bed Reactor
SS	Suspended Solids
SW	Softwood
TMP	Thermo-Mechanical Pulp
TOC	Total Organic Carbon
TS	Total Solids
UASB	Upflow Anaerobic Sludge Blanket
UF	Ultrafiltration
UV	Ultraviolet
WO	Wet Oxidation

the most important barriers. Although European Commission [6] has described the best available technologies to be adopted in large scale applications by pulp and paper mill industry, there is still a need for the industry to continue the collaborations with the researchers to prove and implement new emerging technologies for higher water quality standards. This paper aims to explain the role of chemical additives in wastewater treatment, and also to address the chemical free methods, specifically for the treatment of the P&P mill effluents. The novel treatment methods which have been developed very recently, have also been critically reviewed based on their individual role and their cumulative effects when combined with biological or other physical-chemical methods.

2. Novel P&P mill wastewater treatments

As stated before, it is of high importance to evaluate each treatment method according to several criteria including their treatment efficiency, their subsequent environmental drawbacks and also the economic issues attributed to the treatment method under consideration. Accordingly, this section has been divided into three main topics involving methods with the application of chemical additives (Section 2.1), non-additive methods (Section 2.2) and a combination of both mentioned treatment methods (Section 2.3) to deal with the effluents from pulp and paper mill industries.

2.1. Additive-based treatment methods

The addition of various chemicals (such as coagulants, oxidation agents, etc.) has been developed as a promising way to treat highly polluted and complex effluents. This section considers the recently developed methods based either on the addition of a single chemical, or those relying on the addition of multiple chemical compounds for the treatment of pulp and paper mill effluents. Considering that the sustainable development of the treatment methods are highly related to both the efficiency of the applied method and also to the probable subsequent toxic effects of the chemical additives, such features have been highlighted in this section [7].

2.1.1. Single-additive treatment methods

The addition of a single chemical to the content of the pulp and paper mill effluents is the basis of conventional treatment methods such as adsorption, flocculation and coagulation, oxidation, etc. However, recent published papers have mainly focused on the adsorption and oxidation when just a single strategy is desired for the treatment of pulp and paper mill effluents.

Adsorption has been demonstrated as an efficient method for the treatment of various types of wastewaters due to numerous advantages such as simplicity in design as well as cost-effectiveness in terms of initial investment and land requirements [8]. In order to remove both colour and organic/inorganic contaminants from pulp and paper mill wastewater, various adsorbents including activated carbon, silica, fuller's earth, coal ash, bentonite, etc. have been applied [1]. Among the existing adsorbents, it has been reported in the literature that activated carbons can offer a higher adsorption performance due to its specific characteristics such as high specific surface area, large pore volume, high-speed kinetic, and coarse texture [9,10]. However, drawbacks such as being expensive and the need for costly and time-consuming measures to remove the molecules adsorbed inside the pores [11] have stimulate the search to explore substitutes that are available, with low-cost, are reusable and ease of application [12].

Over the recent years, the performance of various adsorbents has been tested in order to remove dyes and other pollutants from PPMW. Sajab et al. [10] studied the effects of oil palm empty fruit bunch (EFB) fibers modified with cationic polyethylenimine on the removal of colour and organic pollutants from bleaching P&P effluents originated from two treatment stages: primary clarifier and biological aeration tank, named as A and B effluents, respectively. Their experimental results indicated that by increasing the adsorbent dosage, colour and biological oxygen demand (BOD) were significantly reduced in both A and B effluents. For the adsorbent dosage of 9 g/L, there was a BOD_5 reduction of approximately 32.3% and 90.4% in A and B effluents, respectively. Moreover, the maximum colour removal was reported to be 93.6% and 87.5%, respectively, although total organic carbon (TOC) was only slightly reduced. It should be also noted that in their study, an inverse relationship was observed between pH and colour and TOC removals.

As discussed by Kamali et al. [5] some effluents, especially those from *Eucalyptus* sp. pulp and paper making processes, may contain relatively high amounts of phosphorus. In this case, cost effective methods are also required to treat such effluents preventing their discharge to the environment. Among the recent studies for the removal of phosphorus from the effluents, Barbosa et al. [13] suggested the application of ashes (both fly and bottom ashes [14]), because of their high phosphorus adsorption capacity and their low cost. By addition of fly ash or bottom ash to a P&P mill wastewater (solid/liquid ratio = 34.45 g/L for fly ash and 46.59 g/L for bottom ash) more than 90% of the phosphorus removal was achieved. Ecotoxicological studies using two organisms including *Vibrio fischeri* and *Artemia franciscana* also indicated that there was no acute toxic effects of the supernatant effluents resulting from treated P&P mill wastewater using this method. Although there are some reports in the literature on the application of ashes from various origins for the treatment of different effluents, as a single treatment additive [15,16], the number of published works on the treatment of effluents from P&P mills using these materials is rare. It is worthy to note that this technique can provide an opportunity to deal with highly polluted effluents such as those from P&P mills, especially those from bleaching stages.

Providing adequate nitrogen sources for the microorganisms has also been an issue for conventional wastewater treatment plants using biological processes. So, exploring proper nitrogen sources as an additive to enhance the microbial activities would be a need for most wastewater treatment practices. Blank et al. [17] applied a tertiary treatment by controlling the algal growth inside a photobioreactor. They indicated chitin as a good alternative source of nitrogen for the treatment of P&P mill wastewater using algae. Their results showed that algae and cyanobacteria can grow well in the presence of chitin, and they can uptake and remove the phosphorus from the effluent.

The application of wastes from treatment plants for the treatment of industrial effluents can be an attractive candidate, especially in terms of economic considerations. As an example, a batch adsorption system was developed by Khan et al. [18] in which the inactivated secondary excess sludge obtained from the treatment of P&P mill effluent was utilized as an adsorbent to remove three major phytosterols: ß-sitosterol, ß-sitostanol and campesterol during secondary PPMW treatment. In their study, increasing the concentration of inactivated secondary excess sludge from 20 to 2000 mg/L caused more than 80% reduction in liquid-phase sterol concentration during the first 2–4 h. Moreover, following the same trend, all the three mentioned plant sterols were easily adsorbed onto the inactivated sludge.

Surface area is a main characteristic for an adsorbent as it may determine its available active surface sites to adsorb the targeted pollutants. Hence, the interest of very recent studies has also been to maximize the surface area of the adsorbents. Boonpoke [9] examined the effectiveness of adsorption method using water hyacinth based-activated carbon with high surface area (1066 m^2/g) in both batch and continuous experiments for PPMW treatment. Within the first 40 min, chemical oxygen demand (COD) and colour were removed rapidly. The highest removal efficiency for COD and colour (91.70% and 92.62%, respectively) was observed under continuous mode. Furthermore, it can be an area for further studies to develop adsorbents with very high specific surface area using some novel techniques in the preparation of these materials e.g., ultrasonic irradiation [19,20] in order to prepare adsorbents with both high surface area and high adsorption capacity [21,22].

The use of ion exchange resins for adsorbing harmful contaminants from P&P mill effluents has also became recently of interest to industries. In this regard, polystyrene spheres (PSs) have been extensively applied as a matrix substance due to its advantages such as cheapness, availability and high mechanical stability. However, the cationic or anionic groups on the surface of conventional ion exchange resins are of short-molecular chains [23]. In order to overcome this problem, Xiao et al. [24] through the application of modifications to the spheres surface with a fibrous polymer, intended to study the efficiency of cationic polystyrene spheres for the removal of anionic contaminants from paper-making white water. The process started with the acylation of PS spheres by acryloyl chloride. Then, poly methacryloxyethyl-trimethyl ammonium chloride (PMAC) was grafted onto cationic PSs spheres using surface-initiated free-radical polymerization approach. The effect of temperature and reaction time on the removal of dissolved and colloidal substances (DCS) was then investigated in the cationic PSs concentration of 10 g/L. Their results showed that the adsorption performance of cationic PSs can be improved by increasing the temperature due to raising the molecular motion in the system. Furthermore, at longer reaction times, more DCS could be adsorbed onto the surface of cationic polystyrene spheres. Finally, the obtained cationic PSs spheres indicated the high reusability feature which can be considered as a cost-effective adsorbent for reducing the DCS from the white water.

Reusability studies have also been addressed in very recent studies. Mixing adsorbent materials such as biochar, organobentonite and activated carbon with zero-valent iron (ZVI) particles was found to be a beneficial way for separating the adsorbate from the adsorbent, which resulted in the increase of the adsorbent life time [25]. In addition, it has been reported in the literature that doping adsorbents with another compound can enhance their overall efficiency. For instance, doping with some metal catalyst like nickel can enhance the dechlorination rate of pentachlorophenol (PCP) by zero-valent iron magnetic biochar composites (ZVI-MBC). The removal efficiency of Ni-ZVI-MBC for the PCP removal from the synthetic and real PPMW was evaluated by Devi and Saroha [26]. They reported that Ni-ZVI-MBC is the most efficient compound among the studied materials (97.5% in 60 min). However, the application of advanced adsorbents such as biopolymers [27] and advanced nanomaterials [27] can be recommended for further studies in order to develop effective and cost-benefit methods to deal with highly polluted and complex wastewaters from P&P industry.

There are several reports in the literature on the application of oxidation processes for the treatment of PPMW effluents [4]. However, it can be stated that the full-scale implementation of the most conventional oxidation methods is not economically feasible due to high energy and chemicals requirements for the complete degradation of recalcitrant compounds [28]. Hence, advanced oxidation processes (AOPs) which are able to convert the recalcitrant compounds into more biodegradable substances are considered as a promising alternative within the existing techniques of the PPMW treatment [29,30]. Among the various AOPs, ozone (e.g., as O_3/UV or O_3/H_2O_2), Fenton's process (H_2O_2/Fe^{+2}), photo-Fenton reactions (UV/H_2O_2/Fe^{+2}), and UV radiation together with hydrogen peroxide (UV/H_2O_2) were found to have good performances for removing the recalcitrant contaminants from P&P mill effluents [31]. In general, according to the results obtained from the relevant studies, ozonation with a COD removal of approximately 40% has been successfully implemented at industrial scale, mainly due to the presence of these type of facilities previously allocated for pulp bleaching in some paper mills, whereas Fenton processes with a better efficiency for COD removal (approximately 65–75%) at a laboratory scale, can be considered an opportunity for further developments in order to be used at large-scale applications [32].

Some recent studies have also been successfully applied, using nano-scale catalysts for the wet oxidation (WO) of PPMW. For example, Anushree et al. [33] investigated the catalytic wet air oxidation (CWAO) of PPMW by NiO-CeO_2 nano-catalysts, prepared via a co-precipitation process. The results showed 62% and 75% of COD and colour removals, respectively, at the mild operational conditions of 90°C and 1 atm. These results can be considered as a proof for the effectiveness of nanomaterials (such as $Ce_{40}Ni_{60}$ nano-catalyst) for the oxidation of non-biodegradable compounds, due to their low size, high surface area and pore size and volume. Anushree et al. [34] applied the synthesized mesoporous $Ce_{1-x}Fe_xO_2$ mixed oxides as heterogeneous catalysts for CWAO of paper mill wastewater. Under optimal conditions (temperature 90 °C, pressure 1 atm, catalyst dosage 1 g/L, pH = 4, and reaction time of 2 h), $Ce_{0.4}Fe_{0.6}O_2$ mixed oxides revealed optimum COD and colour removals (74% and 82%, respectively). In addition, examining the possibility of catalyst recovery and reuse exhibited that $Ce_{0.4}Fe_{0.6}O_2$ can be applied for three times without a significant reduction in catalytic performance. Non-catalytic wet oxidation methods have also been wider applied compared to CWAO approach due to its both operational simplicity and low cost [35,36]. In a study of a non-catalytic WO process, Dudala et al. [37] obtained COD and colour reductions of 55% and 70%, respectively, from synthetic pulping liquor. The reaction was performed for 4 h at 170 °C and 0.37 MPa of pressure. At the end of the oxidation process, the biodegradability of the wastewater was increased from 0.22 to 0.66. From these results, it may be inferred that the non-catalytic wet oxidation process can also present an acceptable efficiency to reduce contaminants from PPMW.

Combination of oxidation processes and electricity is another option to maximize the treatment efficiency of the pulp and paper mill effluents. Amongst the electrochemical advanced oxidation processes (EAOPs), anodic oxidation (AO) method has attracted a great attention over recent years for reducing toxic and recalcitrant compounds from highly polluted and complex wastewaters [38]. In this process, highly reactive hydroxyl radicals (•OH) are generated at the anode surface (M), according to Eq. (1) [39].

$$M + H_2O \rightarrow M(\bullet OH) + H^+ + e^- \quad (1)$$

Anodic oxidation process is an environmentally friendly technique in which various materials such as platinum, graphite, doped and undoped PbO_2, boron-doped diamond electrodes (BDDs), etc. can be used as anodes [40]. In this respect, Salazar et al. [41] treated raw acid and alkaline bleaching effluents from a hardwood-based Kraft pulp mill by AO-H_2O_2 process containing an air-diffusion cathode and a DSA-RuO_2 or BDD anode at constant cell voltage (2–12 V). In their study, BDD anode showed a much better performance in the mineralization of the recalcitrant organic compounds when compared to DSA-RuO_2 anode after 9 h, enhancing TOC removal by 75% and 65% from alkaline and acid effluents, respectively. This can be explained by the higher oxidizing capability of its physisorbed $^{\bullet}OH$ and its more inert surface. Three dimensional electrode reactors are another possible solution in this regard [42–46]. Jing et al. [47] used a novel three-dimensional electrode reactor for the degradation of acidified reed pulp black liquor (Fig. 1). Both types of direct electrolysis based on anodic oxidation and indirect electrolysis based on microcell mechanism revealed a TOC removal percentage of approximately 35.57%. It should be concluded that the three-dimensional electrode systems can be successfully applied for the degradation of PPMW due to their high specific surface area, low energy requirement and high mass transfer when compared to the traditional two-dimensional electrode systems.

The application of polysilicate composite coagulants, as newly developed inorganic polymer coagulants based on poly-silicic acid (PS) and metal salts, has attracted great attention in the wastewater treatment field, due to their distinctive capabilities for charge neutralization, as well as for adsorption and coagulation of colloidal particles [48]. In this regard, He et al. [49] prepared a poly-silicic-cation coagulant with different Si/(Al + Fe) molar ratios through synchronous polymerization, for the tertiary treatment of recycled paper mill wastewater. In this study, the polymeric aluminum chloride (PAC) was used as benchmark with respect due to its high-quality coagulant ability. The results clearly showed a lower cost as well as an excellent performance of the new poly-silicic-cation coagulant for the removal of COD, colour and turbidity (67%, 95%, and 99%, respectively) from the pulp and paper mill effluents when compared with the traditional PAC treatment.

2.1.2. Multiple additive treatment methods

The application of a combination of two or more single additives is another opportunity to enhance the treatment efficiency of the pulp and paper mill effluents due to the synergic effects of the individual additives on the removal of recalcitrant organic pollutants. However, the number of the combined additive methods is relatively rare in the literature for the treatment of pulp and paper mill effluents. Sticky contaminants is a prevalent problem in mills producing pulp and paper from recycled fibers which can lead to serious consequences on the paper machine, on the production downtime due to the increase in the proceedings related to the maintenance, cleaning, replacing of equipment, etc., and on the increase of the production wastes [50]. Liu et al. [51] evaluated the use of a bentonite & polyacrylamide (PAM) particle flocculation system for the treatment of recycled fiber PPMW. Under the optimal conditions (bentonite dosage: 150 mg/L, PAM dosage: 10 mg/L, pH: 6.4, temperature: 60 °C), 91.26%, 90%, 99.56%, and 95.58% of methyl-tert-butyl ether (MTBE) extract, COD, turbidity and suspended solids (SS), respectively, were removed. Additionally, the average particle size after treatment was reduced from 40.95 μm to 0.5 μm. Therefore, the bentonite adsorption and coagulation treatments were found to be efficient for recycled fiber pulp wastewater treatment, particularly due to its high capacity for the removal of large size particles and suspended colloidal substances like stickies from PPMW. There are also recent reports on the application of industrial wastes for the treatment of industrial effluents. For instance, Yang et al. [52] prepared an inorganic polymeric ferric aluminum sulfate chloride (PFASC) composite coagulant using waste pickling liquor from steel mills together with polyacrylamide (PAM) in a tertiary treatment. They removed COD and chroma 65.3% and 71.2% respectively, under initial pH 7.5, 1 mL/L PFASC, 1.0 ppm PAM. Very recently, the application of combined additive systems using advanced oxidation processes have also gained a huge attention. For instance, Gopalakrishnan et al. [53] used solar/Fe^{2+}/TiO_2/H_2O_2 process for the treatment of pulp and paper wastewater and found a maximum COD and colour removals of 98% and 97% respectively at optimum conditions of flow rate = 75 mL/min, liquid depth = 5 cm and residence time = 75 min. AOPs using Fenton reactions have also demonstrated good performance with other physic-chemical methods. Grötzner et al. [54] achieved 95% for TOC, 61% for COD, and 76% for lignin contents removals using a sequence of coagulation-flocculation-sedimentation and AOP by Fenton process for the treatment of chemical thermal mechanical pulping effluents from a Brazilian pulp and paper industry.

As a conclusion, it seems that there is room for further studies and developments in the application of combined low-cost and high efficiency chemical additives. Meanwhile, studies on the toxic effects and the relevant environmental drawbacks and life cycle assessment of the combined additives would be essential to promote the full-scale application of such methods. Table 1 provides a summary of the very recent publications on the use of additives for the treatment of pulp and paper mill effluents.

2.2. Non-additive treatment method

This section aims to review the very recent advances in the treatment of P&P mill effluents using the methods which are independent of chemical additives application. To this end, physical, biological treatments and their combinations are included in this section.

2.2.1. Physical treatments

So far some single physical treatments such as sonochemical irradiation [55], electrical flow [56], or a combination of different physical treatments [57] have been applied for the decontamination of effluents from various origins. However, the number of literature publications on the single physical treatments of PPME is relatively rare. With the aim of reducing colour and turbidity, Shaw and Lee [58] carried out an ultrasonic treatment (at 357 kHz) of effluents from Kraft P&P mill. It was observed that the

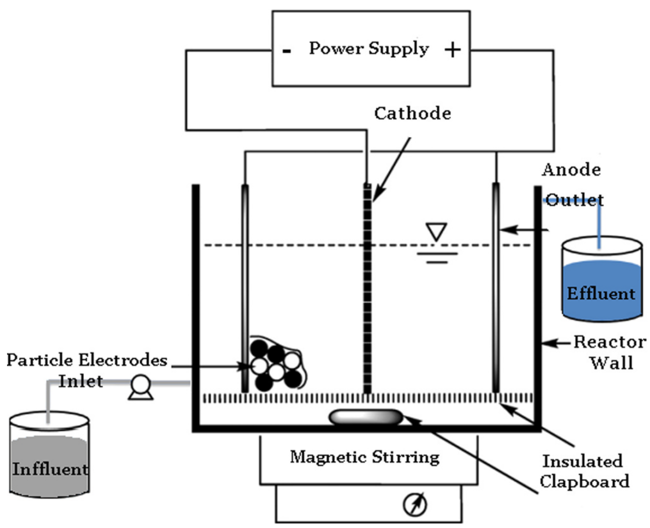

Fig. 1. A schematic of a three-dimensional electrode reactor, adapted from Jing et al. [47].

Table 1
A summary of the very recent additive based methods applied for the treatment of pulp and paper mill wastewater.

Method	Experimental conditions					
	Effluents origin	Time	pH	Additive(s)	Additive dosage (g/L)	Temperature (°C)
Adsorption	Bleaching		7.3	Oil palm EFB	9	
			8.5	Oil palm EFB	9	
Adsorption				Fly ash	34.45	
				Bottom ash	46.59	
Adsorption		Between 2–4 days		Chitin	2	
Adsorption	Sterol spiked PPMW	First 2–4 h	7	PPM Secondary Sludge	2	
Adsorption		40 min		WHAC[a]		
Adsorption	With water	Initial minutes	–	Cationic PS	10	40
Adsorption	Synthetic and real effluents	240 min	6	Ni-ZVI-MBC	0.2	40
CWAO	After primary clarifier	150 min	4	NiO-CeO_2 nano-catalysts	1	90
CWAO	Primary clarifier	120 min	4	Ce0.4Fe0.6O_2 mixed oxides	1	90
Anodic Oxidation	Bleaching	9 h	10.5	BDD anode		
Anodic Oxidation	Bleaching	9 h	2.5	BDD anode		
Anodic Oxidation	Black Liquor	2 h	2.5	GACE[b]		25
Coagulation	Aerobic PPMW		7.2	PSC[c]	< 10	–
Adsorption & Coagulation	Pulping, floatation deinking	2–5 min (stirring)	6.4	Bentonite	0.15	60
				PAM	0.01	

Method	Parameters									Ref.
	COD		BOD		Color (%)		Other parameters			
	Initial (mg/L)	Removal (%)	Initial (mg/L)	Removal (%)	Initial (pt-Co)	Removal (%)	Parameter/ substance	Initial	Removal	
Adsorption	1500		240	32.3	963	93.6				[10]
	122		148	90.4	429	87.5				
Adsorption							Phosphorous		90%	[13]
Adsorption							Phosphorous	280 μg/L	2.6 μg/L day	[17]
Adsorption	736 ±15						Sterol: campe	850 μg/L	80%	[18]
							Sterol: ß-sitosta	2300 μg/L		
							Sterol: ß-sito	3500 μg/L		
Adsorption	674	91.7			831	92.62				[9]
Adsorption									53.8%	[24]
Adsorption							PCP	1.77 mg/L	100%	[26]

(continued on next page)

Table 1 (*continued*)

Method	Parameters									Ref.
	COD		BOD		Color (%)		Other parameters			
	Initial (mg/L)	Removal (%)	Initial (mg/L)	Removal (%)	Initial (pt-Co)	Removal (%)	Parameter/ substance	Initial	Removal	
CWAO	865	62	234		2768	75				[33]
CWAO	865 ± 32.14	74	234 ± 12.84		2768 ± 114.46	82				[34]
Anodic Oxidation	1500 ± 12	–	595 ± 5		1077 ± 3		TOC	594 mg/L	75%	[41]
							AOX	25.1 mg/L		
Anodic Oxidation	1250 ± 12	–	563 ± 6		496 ± 4		TOC	499 mg/L	65%	[41]
							AOX	19.2 mg/L		
Anodic Oxidation	220.6487 ± 6487	–					TOC	83.70 mg/L	35.57%	[47]
Coagulation	120	67				95	Turbidity	70 NTU	99%	[49]
Adsorption & Coagulation	3020	89.67	–	–	867.4	93.08	Turbidity	108 NTU	99.56%	[51]
							MTBE extract	236.8 mg/L	91.26%	
							Particle size	40.95 µm	0.51	
							CD	1065.4 µmol/L	85.92%	
							SS	860 mg/L	95.58%	

[a] Water hyacinth based-activated carbon.
[b] Granular Activated Carbon Electrode.
[c] Poly-silicic-cation with Si/(Al + Fe).

irradiation of ultrasound for 25 h resulted in the drop in the absorbance of the spectra above 250 nm for aromatic compounds. Ultrasound irradiation (20 kHz–10 MHz) can cause the cavitation involving the formation, growth, and instantaneous implosive collapse of bubbles in the liquid. This process generates local hot spots with high temperatures (up to 5000 °C) and pressures exceeding 500 atm [59] together with shock wave and micro-jet (400 km/h) [60,61]. Under these conditions, the destruction of the aromatic compounds in the content of the effluent is facilitated. The simultaneous removal of colour and COD by using physical methods has been reported by Ebrahimi et al. [62]. They developed a multi-stage ceramic membrane system consisting of a microfilter, an ultrafilter and a nanofilter to diminish COD and residual lignin from alkaline bleaching effluent of a P&P industry (Fig. 2). Such a combination can act as an energy-saving and environmentally friendly method for the removal of contaminants and organic matters from PPME as aimed by some other related studies [63–67]. According to the results obtained by Ebrahimi et al. [62], the sequential two-stage process using microfilter and ultrafilter resulted in a good performance in terms of COD and residual lignin removal by 45% and 73%, respectively.

2.2.2. Biological treatment methods

Biological treatment methods, which rely on the activity of various strains of microorganisms to break down organic substances and harmful compounds, seem to be economically and environmentally superior to most physical and chemical methods applied for the treatment of different types of effluents, including PPMW. The main problem with the application of biological treatments has been the incomplete treatment of the stream, especially in the case of the presence of recalcitrant pollutants such as AOXs in bleaching effluents and lignin-derivatives in the liquor. However, some efforts have been very recently applied to reinforce the bacterial activities to remove such complex compounds. Hooda et al., [68] employed a rod-shaped Gram-positive bacterial strain RJH-1 (isolated from sludge) for the treatment of pulp and paper mill effluents. Although during initial 24 h of experiments, no significant changes happened to COD and colour, probably due to the slow adaptation of the bacteria to wastewater conditions, after a 5 days batch experiments 69% COD, 47% colour and 37% lignin removals were observed. It is worthy to note that microbial strain was also capable to reduce AOXs by 39%. Other microbial strains have been also recently extracted from other media and applied for the P&P mill effluents treatment. Tyagi et al. [69] isolated two dominant bacteria *Bacillus subtilis* and *Micrococcus luteus*, as well as one fungi *Phanerochaete crysosporium* from the soils polluted with P&P mill effluent. They applied microbial stains for the treatment of the effluents from a pulp and paper industry and achieved a lignin removal of 97% after 9 days. In this period, the consortium was also capable to remove 87.2% and 94.7% of BOD and COD, respectively. Similar removals of BOD and COD by other bacterial consortiums have also been recently observed. Significant removals of BOD, COD, and total solids (TS) of 81.25%, 53.23%, and 81.19%, respectively, were achieved by Nadaf and Ghosh [70] by utilization of a non-sporulating bacteria, namely *Rhodococcus sp*. NCIM 2891 for the degradation and detoxification of PPMW. Likewise, the concentration of heavy metals recorded from the biologically treated effluent was less compared to that of influents. Besides the bacterial consortiums, some algal communities have also shown a good performance for removing the organic material, toxic compounds, and nutrients from PPMW in a cost-effective manner [71]. For example, Usha et al. [72] reported removal efficiencies of 82%, 75%, 65% and 71.29% for BOD, COD, NO_3-N and PO_4-P, respectively, from PPMW using a mixed culture of microalgae in an outdoor open pond.

Natural-like systems such as constructed wetlands (CWs) have also recently attracted a significant interest as promising biological options to diminish the nutrients and the pollutants from wastewaters [73]. The main advantages of such systems are their cost-effectiveness and less environmental impacts as compared to conventional treatment approaches [74]. Choudhary et al. [75] studied the performance of horizontal subsurface flow (HSSF) constructed wetland planted with *Canna indica* for the removal of pollutants from PPMW. At an HRT of 5.9 days, the average percentage removals of 89.1% and 67–100% were achieved for AOX and chlorophenolics, respectively, from PPMW. Moreover, Arivoli et al. [76] evaluated the efficiency of vertical flow constructed wetlands by using three common aquatic macrophytes (i.e. *Typha angustifolia*, *Phragmites australis* and *Erianthus arundinaceus*) for the removal of

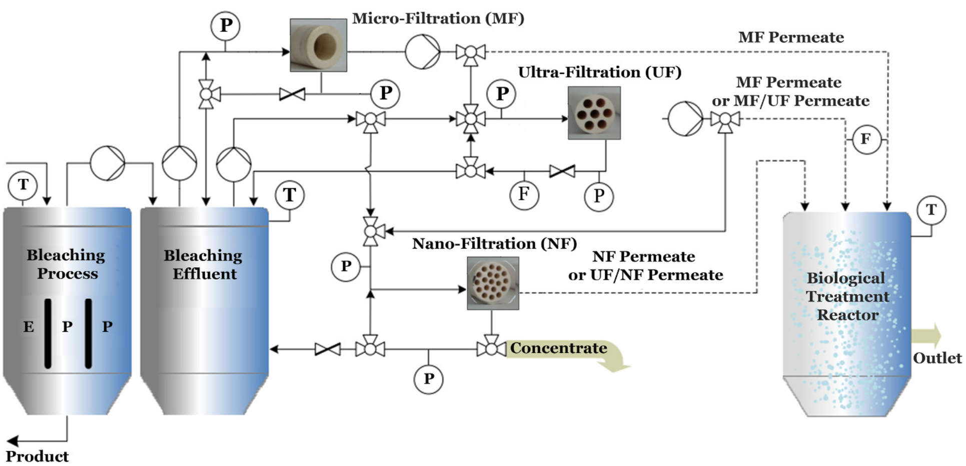

Fig. 2. A schematic of a multi stage filtration process for the treatment of bleaching effluents, adapted from Ebrahimi et al. [62].

heavy metals from P&P mill effluent. The results showed average removals of 74%, 80%, 60%, 70%, 71%, and 70% for iron, copper, manganese, zinc, nickel, and cadmium, respectively. Furthermore, among the three species of macrophytes used in CWs, *E. arundinaceus* showed the best performance.

Anaerobic digestion (AD) processes with different levels of performance have been also reported in the literature for the bio-treatment of P&P mill effluents. Nevertheless, among the various kinds of effluent streams generated from P&P mills, the full-scale AD has been only performed successfully for effluents from the bleached/unbleached thermo-mechanical pulp (TMP) and chemical thermo-mechanical pulp (CTMP) as well as neutral sulfite semi-chemical (NSSC) and Kraft/sulfite mill condensates [77]. It is worth noting that despite AD has numerous advantages, especially low excess solid wastes produced and energy recovery in the form of biogas, the existence of some obstacles has restricted the full-scale applications of AD in the P&P mills. These barriers are mainly related with the innate hardness in associated to the digestion of non-biodegradable compounds such as lignocellulosic material and also facing with a diversity of inhibitors (e.g., sulfur, resin acids, ammonia, heavy metals, and organochlorine compounds) as well as the necessity of adjusting specific operational conditions and the need for adopting proper reactor configuration for anaerobic degradation optimization [78]. Very recent studies have been mainly focused on overcoming these obstacles and optimizing the AD performance for complying with the stringent environmental standards and achieving economic benefits. In an effort, Steffen et al. [79] studied an anaerobic digestion experiment for evaluating the biochemical methane potential (BMP) of separating fines from industrial recycled fiber pulp (RFP). In order to obtain a better understanding, conventional chemical and mechanical pulps were used as benchmarks for methane production. While methane yields of fines from mechanical pulps and RFP were measured as 21–28 mL/g_{VS} and 127 mL/$g_{VS,}$ respectively, a highest methane yield (375 mL/g_{VS}) was attained for fines fractions from chemical pulps owing to the fact that refining chemical pulp increases the fibers' bonding surface area, thereby causing greater access of microorganisms to the fibers and increasing rate of biodegradation. On the contrary, a large amount of lignin in the fines fractions from mechanical pulps, in addition to a high content of inorganic fine particles ($CaCO_3$) and the greater hydrolysis residue regarding the separated fines from RFP might be the reasons for lower gas production. The results suggested that the fines fractions from RFP are appropriate for biogas production even though the obtained biogas yields were merely one-third as compared to the ones obtained with chemical pulp fines.

Amongst the various types of anaerobic reactors, upflow anaerobic sludge blanket (UASB) has retained a special place in the P&P industry over the past few years. Indeed the main reasons for adopting this type of reactor are related to its profitability in terms of high organic loading rate (OLR) capacity, together with the low operation expense, and the production of rich methane biogas as by-product [80,81].

When focusing on the anaerobic treatment of especial P&P mill effluents, some opportunities for further studies are still identified. For instance, very few studies have been carried out so far on the treatment of bagasse effluents from P&P industry especially by conventional AD reactors such as UASBs [82]. Furthermore, no model has been developed to predict the COD removal percentage, COD removal rate, and biogas production from the mentioned effluents. Sridhar et al. [83] studied the anaerobic digestion of bagasse effluents from P&P industry to explore the interactive effects of influent chemical oxygen demand (COD_{in}), hydraulic retention time (HRT), and temperature on the performance of a continuous UASB reactor. In their work, the Box-Behnken design was applied for analysis and modeling the interactive effects of the three variables (COD_{in}, HRT, and temperature) on the responses. Based on the obtained results, under optimum conditions (COD_{in}: 6212 mg/L, HRT: 23 h, and temperature: 35 °C), the highest values of COD removal percentage (84.3%), COD removal rate (230.9 mg/L h), and biogas production (21.2 l/d) were achieved.

Operating a UASB reactor for anaerobic treatment of a bleaching stream can lead to more satisfactory results than the total effluents generated from a P&P mill; as bleaching streams typically contain a higher concentration of organic substances or, in other words lower volumes of water need to be treated to achieve the same results. Larsson et al. [84] investigated the performance of two lab-scale UASB reactors working in *meso*philic condition on the treatment of two types of alkaline kraft elemental chlorine-free bleaching wastewaters (hardwood and softwood). The results showed the filtered total organic carbon (fTOC) reduction of 43% and 60% as well as the biogas production of 120 NmL g $TOC_{IN}{}^{-1}$ and 250 NmL g $TOC_{IN}{}^{-1}$ for softwood (SW) and hardwood (HW) wastewaters, respectively. The methane content of the produced biogas was 75% for both reactors. Based on the results achieved, UASB application can be considered as an appropriate technique for anaerobic digestion of alkaline bleaching effluents produced in kraft PPMs.

Development of novel and advanced types of anaerobic reactors has also been a topic of high interest in recent studies. Some types of AD reactors such as static granular bed reactor (SGBR) without the need for mixing, and aiming to save energy, have been a subject of interest, but still at a lab-scale application [85]. Turkdogan et al. [86] compared the performance of a SGBR with a UASB reactor for the treatment of thermo-mechanical paper mill wastewater in a laboratory-scale experiment. This study was performed within 110 days with hydraulic retention times (HRTs) of 4, 6, 9 and 24 h. The results indicated that in both reactor systems the COD removal efficiency was improved with the increase in HRT, ranging from 67% to 92% for SGBR and 60–90% for the UASB reactor. Thus, SGBR exhibited not only exhibited approximately identical ability as UASB for the removal of organics from P&P mill wastewater, but was also slightly better than the UASB in terms of operational difficulties. So, SGBR system would be a proper alternative as conventional reactors for the anaerobic digestion of P&P mill effluents. Another type of anaerobic reactor applied to the treatment of P&P mills wastewater is the moving bed biofilm reactor (MBBR) which can result in an acceptable removal of organic matter and also in the biosynthesis of added-value products [87,88]. For example, Baeza et al. [89] treated a real PPMW by MBBR, aiming to evaluate the effect of different operational parameters (i.e. BOD_5/nitrogen (N)/phosphorus (P) ratio) on the Polyhydroxyalkanoate (PHA) biosynthesis rate during two experimental phases. In phase I, the best results for the accumulation of PHA (85.10%) and removal of organic matter (95.60%) were obtained using a BOD_5/N/P ratio of 100:5:1. However, for phase II, the highest PHA percentage (89.41%) and organic removal (97.10%) were achieved with BOD_5/N/P ratios of 100:1:0.3 and 100:5:1, respectively. The

Table 2
A summary of the very recent non-additive based methods applied for the treatment of pulp and paper mill wastewater.

Method	Experimental conditions					Production efficiency			
	Effluents origin	Time	pH	Other conditions		Gaseous products	Value	Other products	
				Parameter	Value				Value (%)
Membrane (microfiltration followed by ultrafiltration)	Alkaline bleaching		10.65	Tem.	60 °C				
Microalgae cultivation		28 days	5.41						
Bacterial treatment			4.5						
HSSF constructed wetland		5.9 days	7.7						
Vertical flow constructed wetlands			7.80						
UASB	Bagasse	23 h	4.5–5.5	Tem.	35 °C	Biogas	21.2 l/d		
UASB	Alkaline kraft elemental chlorine-free bleaching (SW)		8			Biogas	120 NmL g $TOCIN^{-1}$		
	Alkaline kraft elemental chlorine-free bleaching (HW)					Biogas	250 NmL g $TOCIN^{-1}$		
SGBR	Thermo-mechanical	4–24 h	7.14	Tem.	35 °C				
UASB									
MBBR	Pressure ground wood pulping		6.83	OLR	2.99 kg BOD_5 m^{-3} day^{-1}			Polyhydroxyalkanoate	85.10
				BOD_5/N/P	100:5:1				
	Thermo mechanical pulping		6.77	OLR	2.83 kg BOD_5 m^{-3} day^{-1}			Polyhydroxyalkanoate	89.41
				BOD_5/N/P	100:1:0.3				
AG-SBR with additional biomass	Dissolved air flotation recycled PPMW	24 h	8						

(continued on next page)

Table 2 (*continued*)

Method	Experimental conditions					Production efficiency			
	Effluents origin	Time	pH	Other conditions		Gaseous products	Value	Other products	
				Parameter	Value				Value (%)
Microalgae cultivation	Mixtures of PPMW with dairy wastewater		7.37	Tem.	27.5 °C			Biomass yield	> 37
Photo fermentation	PPMW with POME (3:1)	72 h	7.00	Tem.	30 °C	Biohydrogen yield	4.670 mL H_2/mL		
Hybrid UFBAB reactor and SSF		24 h	8.9						
UI and Photo fermentation		45 min	6.9	Tem.	30 °C	Biohydrogen yield	0.041–0.077 mL H_2/mL		
				UI[a] amplitude	60				
UI and Photo fermentation	Mixtures of PPMW with POME (3:1)	45 min	7.00	Tem.	30 °C	Biohydrogen yield	8.72 mL H_2/mL		
				UIamplitude	70		medium		
UI and Photo fermentation		10 min	6.9	Tem.	30 °C	Biohydrogen yield	9.62 mL H_2/mL		
				UI amplitude	30				
		15 min		Tem.	30 °C				
				UI amplitude	15				
Bacterial treatment	Synthetic PPMW	5 days	7	Tem.	37 °C				
Bacterial and fungi treatment		9 days	9	Tem.	26 °C				

Removal efficiency of parameters/substances							Ref.
COD_{total}		BOD		Other parameters			
Initial (mg/L)	Removal (%)	Initial (mg/L)	Removal (%)	Parameter/ substance	Initial	Removal (%)	
10400	35–45			TOC	4000 (mg/L)		[62]
				Lignin		60–73	
3000.15	75	2944	82	NO_3-N	9.932 (mg/L)	65	[72]
				PO_4-P	30.25 (mg/L)	71.29	
7768	53.23	160	81.25	TS	39.33 (mg/L)	81.19	[70]
				Cu	0.12 (mg/L)	16.67	
				Cd	0.05 (mg/L)	40	
				Fe	3.96 (mg/L)	39.14	
				Cr	0.57 (mg/L)	77.19	
				Ni	0.33 (mg/L)	39.39	
1011	87.86	248	93.14	AOX	16.5 (mg/L)	89.1	[75]
				Chlorophenolics	40 (mg/L)	67–100	
				Colour	2553 (Pt-Co)	96.05	

(*continued on next page*)

Table 2 (*continued*)

Removal efficiency of parameters/substances							Ref.
COD_{total}		BOD		Other parameters			
Initial (mg/L)	Removal (%)	Initial (mg/L)	Removal (%)	Parameter/ substance	Initial	Removal (%)	
981.7	55.38	230.2	45.35	Fe	1.56 (mg/L)	74	[76]
				Cu	0.246 (mg/L)	80	
				Mn	0.21 (mg/L)	60	
				Zn	0.39 (mg/L)	70	
				Ni	0.06 (mg/L)	71	
				Cd	0.013 (mg/L)	70	
				Phenol	4.93 (mg/L)	43.20	
6212	84.3						[83]
				fTOC	6300 (mg/L)	43	[84]
				fTOC	610 (mg/L)	60	
1133.9	67–92						[86]
	60–90						
839		441	95.60				[89]
2970		1670	97.10				
1057	95%			Colour	121 (Pt-Co)	82	[91]
				Turbidity	735 (NTU)	95	
				SS	149 (mg/L)	92	
				NH_3-N	4.1 (mg/L)	86	
				PO_4^3-P	0.03 (mg/L)	60	
1905	92.7			NH_4-N	22.35 (mg/L)	99	[96]
				NO_3-N	1.06 (mg/L)	25–55	
				PO_4-P	10.1 1 (mg/L)	> 95	
22 900	28.8						[99]
2912	99	1280	90	Sulfate	207 (mg/L)	> 50	[101]
				Phenols	2.3 (mg/L)	> 70	
				TDS	1270 (mg/L)	66.5	
				Chloride	365 (mg/L)	> 50	
1441	No change			SCOD	318 (mg/L)	25.9	[102]
22,900	36.9			SCOD	5725 (mg/L)	34.2	[103]
1441							[104]
				SCOD	318 (mg/L)	18	
500	69			Colour	1000 (Pt-Co)	47	[68]
				lignin		37	
				AOXs	15(mg/L)	39	
935–1005	94.7	130–136	87.2	Lignin	1.958(mg/L)	97	[69]

[a] . Ultrasonication.

results achieve can emphasize the efficiency of the membrane bioreactors to deal with highly polluted effluents.

Sequencing batch reactors (SBRs) are also among the suitable biological systems for the treatment of wastewaters such as PPME, due to their design simplicity and functional flexibility [90]. Muhamad et al. [91] treated a real recycled paper mill effluent using three forms of lab-scale aerobic bioreactors, containing two attached-growth SBR (AG-SBR) systems with or without additional biomass, and a suspended growth SBR (SG-SBR). Over a 300-days observation period, the attached-growth SBR with additional biomass showed the best operational stability and average removal performance of 95%, 82%, 95%, 92%, 86%, and 60% for the relevant parameters, namely COD, colour, turbidity, SS, NH_3-N, and PO_4^{3}-P, respectively.

The manipulation of anaerobic digestion reactors can also enhance biogas production and treatment efficiency for a given industrial effluent. The main problem here is that in some cases, the effluents have high COD (e.g., in case of black liquor) or high concentrations of toxic materials (e.g., AOXs). As a promising strategy, co-treatment approach can encompass many potential benefits including dilution of inhibitory and toxic compounds, establishing a balance between nutrients as well as increasing the diversity and synergistic effect of microorganisms in the system. Also, economic benefits may be obtained from equipment sharing [92,93]. However, despite the numerous advantages of co-treatment strategies, nowadays very few facilities have been adopted for the process due to some concerns such as the need for upgrading or strengthening the existing facilities, process instability, unsuccessful operation arising from undesirable suspended impurities, and digester overloading which can lead to foaming problems and unexpected failure of the process [94,95]. According to the above shortcomings, so far only few studies have been conducted on the use of co-treatments in P&P industry. Most of the published work have concentrated their attention on the combination of P&P primary and secondary sludge with substrates such as food waste, municipal sewage sludge and monosodium glutamate waste liquor [1]. Co-treatment with microalga is another candidate to enhance the overall performance of the anaerobic digestion process. Gentili [96] mixed the effluents from municipal and industrial sources. Afterwards, three microalgal strains (*Scenedesmus dimorphus*, *Selenastrum minutum* and *Scenedesmussp.*) were grown in three wastewater mixtures (P&P influent 4:1 dairy sludge; P&P influent 1:1 municipal influent; P&P influent 2:1 dairy final effluent), named as a, b and c wastewaters, respectively. Based on the results, the highest lipids yield, up to 37% of the dry matter was obtained in the wastewater mixture (a) by *Selenastrum minutum*. In addition, the highest COD removal (92.7%) was attained in the wastewater mixture (a) with the *Scenedesmus sp*. Therefore, mixing the P&P wastewater with dairy and municipal wastewaters was found effective not only in wastewater treatment but also for the production of biomass with high amounts of lipids.

Photofermentation process, as one of the most desirable biological approaches for biohydrogen production from organic substrates [97], is another option to deal with P&P mill effluents. However, the practical applicability of this approach for lignocellulosic-based wastewaters, such as PPMW, has been restricted with a degradation deficiency and low hydrogen yield due to their complicated and resistant structure to microbial enzymatic attacks [98]. Budiman et al. [99] investigated the potential of co-treatment approach by combining palm oil mill wastewater (POMW) and P&P mill effluent for bacterial growth and biohydrogen production using photofermentation. The results revealed that with a mixture of wastewater containing 25% (v/v) POME and 75% (v/v) PPME the maximum biohydrogen yield of 4.670 mL H_2/mL can be achieved. Moreover, in this study the highest COD_{total} removal (up to 28.8%) was observed during 72 h of photofermentation using *R. sphaeroides* NCIMB8253. Thus, co-treatment of POME with PPME was found to be effective for reducing turbidity as well as for improving the visibility of wastewaters during photofermentation process. Furthermore, by mixing a proper ratio (9:1) of PPME and brewery wastewater (BW), Hay et al., [100] achieved a biohydrogen yield of 0.69 mol H_2/L_{medium} from photofermentation process. This combination also led to SCOD removal, soluble carbohydrate consumption, and light efficiency of 36.7%, 32.1%, and 1.97%, respectively. Table 2 presents a summary of the very recent publications on non-additives processes for the treatment of pulp and paper mill effluents.

2.2.3. Combined physico-biological treatment methods

Due to the insufficient treatment output of the single treatment methods, various studies have claimed the necessity of combining biological and physico-chemical methods [1,5]. Considering the benefits of such combinations, either in terms of economical-environmental aspects as well as the potential synergistic effects, Mishra et al. [101] studied the efficiency of an integrated system including upflow fixed-bed anaerobic bioreactor (UFBAB) and slow sand filter (SSF) for the removal of pollutants from PPMW (Fig. 3) as compared to a single UFBAB. At different HRTs, the hybrid system showed a very high removal efficiency of 90%, 99%, 100%, and

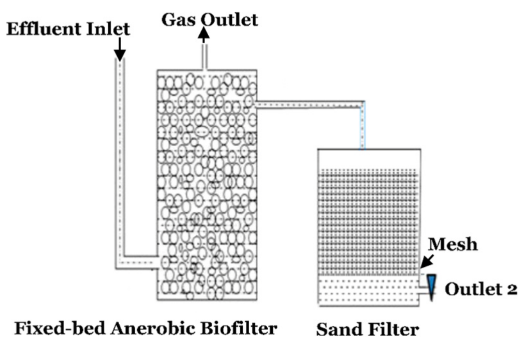

Fig. 3. A scheme of a biofilter designed for P&P mill effluent treatment, adopted from Mishra et al. [101].

100% for BOD, COD and total dissolved solids and total suspended solids, respectively. Similarly, in the case of sulfate and phenols removal, a relative high efficiency was attained, indicating a very good performance for such combined system.

Recent studies have also demonstrated that pre-treatment of lignocellulosic materials by moderate ultrasound irradiation could break down their complex composition via the development and collapse of cavitation bubbles, which can lead to a positive effect on the biohydrogen production from the fermentation process [105,106]. In this regard, Hay et al. [102] investigated the effects of ultrasonic power (30–90%) and duration (15–60 min) on the photofermentation process of PPMW. Based on the results obtained, the biohydrogen yield of all PPMWs pretreated by ultrasonication was higher than the ones obtained with the raw substrate, ranging between 0.041 and 0.077 mL H_2/mL. The maximum cumulative biohydrogen production (430%) and SCOD removal efficiency (25.9%) were achieved at the amplitude of 60% and time of 45 min (A60:T45), respectively. Moreover, economic assessment conducted in this research showed that ultrasonic pretreatment prior to biofermentation of PPMW could have a positive net saving in comparison with raw PPMW, due to the higher incomes from selling the biohydrogen and further reduction in costs related to the COD removal. Once more, Hay et al. [104] applied various ultrasound irradiation times (5, 10 and 15 min) and amplitudes (15%, 30%, and 45%) to specify the optimal ultrasonication conditions for pretreatment of *Rhodobacter sphaeroides*. As indicated in Fig. 4(a) moderate ultrasonic pretreatment could stimulate the generation of pores in the cell membrane of *R. sphaeroides* or slightly disrupt surface of cell, whilst higher-frequency ultrasonic was able to create more pores in cell surface, thereby resulting in further change of the cell morphology as compared to the untreated bacteria cell. Thus, highest hydrogen yield (9.62 mL bioH_2/mL) was attained at A30:T10 due to the fact that moderate ultrasonic applied to *R. sphaeroides* could improve the enzymatic activities of the cell with modification of membrane permeability and resistance, leading to increase degradation of large-size molecules into smaller ones. However, the higher release of intracellular products in the ultrasonic-pretreated PPMW caused a lower SCOD removal efficiency after biohydrogen production process as compared to the control substrate.

In another effort to improve the biohydrogen production, Budiman and Wu [103] evaluated the performance of ultrasound pretreatment (amplitudes of 30–90% and duration of 5–60 min) on a combined substrate consisting of PPMW and POME. The maximum hydrogen production rate (8.72 mL H_2/mL_{medium}) and COD removal efficiency (36.9%) were achieved by using A70:T45 ultrasonication. The improvement of biohydrogen production was linked to the positive effect of ultrasound irradiation on the release of lignin and cellulose from the structure of substrates, leading to increased bioavailable nutrients which could be simply degraded by fermentation bacteria.

2.3. Combined additive and non-additive treatment methods

2.3.1. Physico-chemical combined treatment methods

Application of various physical stimulates such as vibration, electricity flow, temperature, photo irradiation, filtration, and pressure, etc. can potentially enhance the performance of methods based on the addition of chemicals for the treatment of industrial effluents. For instance, the combination of ultrasonic irradiation with AOPs can be an interesting solution for the removal of recalcitrant pollutants from P&P mill effluents. Generation of powerful hydroxyl groups as a result of ultrasonic irradiation can facilitate the oxidation of complex organic materials especially when combined with methods such as Fenton-like oxidation treatments. Eskelinen et al. [107] studied the treatment of bleaching effluents from a P&P mill using a combination of ultrasonic irradiation in combination with Fenton-like oxidation (Fe^{3+}/H_2O_2) or photo-Fenton degradation (Fe^{2+}/H_2O_2/UV). Such an integration resulted in COD removals of 12%, 20%, and 28% from effluents by using Fenton-like oxidation, photo-Fenton's oxidation or electro-oxidation treatment, respectively, at the reaction conditions of dose of Fe(III)-1 g/L; dose of H_2O_2-3 g/L, pH-6.9, agitation speed: 200 rpm, and contact time-60 min. It has been also reported that ultrasonic irradiation can facilitate the coagulation process especially when combined with electricity. Asaithambi et al. [108] showed that the application of sono-electrocoagulation can yield 95% and 100%, for the removal of COD and colour, respectively, at a current density of 4 A/dm^2, electrolyte concentration of 4 g/L, initial pH of 7, COD concentration of 3000 mg/L, electrode combination of Fe/Fe, inter-electrode distance of 1 cm, and reaction time of 4 h. Although filtration is not able to treat highly polluted industrial effluents, as in the case of PPMW, its combination with methods based on the addition of chemical additives can be considered as a good candidate to promote the overall efficiency of wastewater treatment plants. For instance, Gholami et al. [109] applied a hybrid system assembled with ultrafiltration (UF) membrane and AOPs in order to treat an effluent from fiber sewer collection unit. They indicated that the UF permeate quality was not sufficient to satisfy the related environmental regulations. As the next step, they applied AOPs under which sulfate and hydroxyl radicals were generated through the activation of persulfate ($S_2O_8^{2-}$) and H_2O_2 by Fe(II) to deal with the pollutants. Under optimized operational conditions,

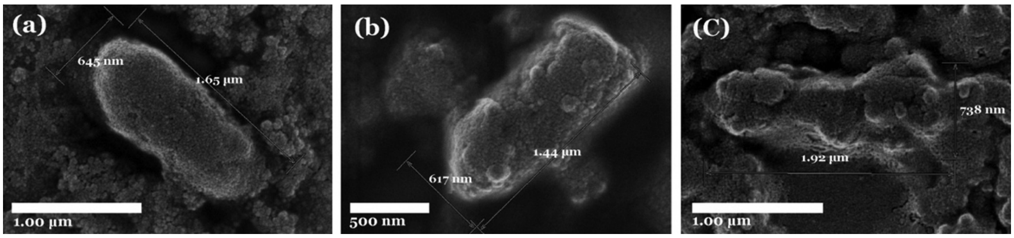

Fig. 4. FE-SEM images of R. sphaeroides NCIMB8253 after undergoing (a) no ultrasonication; (b) ultrasonication with amplitude 30% and 10 min; (c) ultrasonication with amplitude 45% and 15 min, adopted from Hay et al. [104].

($[H_2O_2]$ = 15 mM, [Fe (II)] = 6 mM, and pH = 3), the removal efficiency of COD, UV_{254}, and UV_{280} were 95.02%, 86.74% and 87.08%, respectively. Additionally, in order to reduce the reaction time and costs, Salazar et al. [41] combined AO-H_2O_2 with UF, nanofiltration (NF) and reverse osmosis (RO) treatments. The results for TOC removal by integrated filtration/AO-H_2O_2 indicated that the best pair-treatments for acid and alkaline bleaching effluents were UF/AO-H_2O_2 (68% of TOC removal) and NF/AO-H_2O_2 (96% of TOC removal), respectively. AOPs can also compensate the deficiency of electro-coagulation for the complete treatment of PPMW. Jaafarzadeh et al. [110] mixed electro-coagulation with UV oxidation for the removal of organic compounds from a P&P mill wastewater (COD = 1537 mg/L). Under optimum conditions (natural pH, time = 33.7 min and current density = 5.55 mA/cm^2) they achieved 61% of the COD removal. In a recent study, Abedinzadeh, Shariat and Masoud [111] applied an SBR method in combination with AOPs for the treatment of P&P wastewaters. Using a response surface methodology (RSM), they achieved an optimum COD and colour removals of 98% and 94%, respectively, under Fe^{2+} and H_2O_2 dosages of 3 mM and 9 mM, respectively at pH of 3.0 and 30 min.

For a sustainable treatment of industrial effluents, there is also a need for cheap and environmentally friendly additives. So far, some attempts have been made in this regard. As shown by Zhuang et al., [112] who used waste rice straw and iron-containing sludge to prepare catalytic particle electrodes. Electro-Fenton (EF) oxidation applied to real paper mill wastewater using the prepared electrodes under optimized treatment conditions (current density of 10 mA/cm^2, a catalytic particle electrodes dosage of 1.0 g/L, and an aeration rate of 5 L/min) reached allowed 86% of SCOD removal.

Due to the fact that sun is a renewable energy source that is free but dependent on the season and geographical conditions, it can be used as a source of light for treatment processes. It can also be combined with other physico-chemical methods to enhance the overall treatment efficiency. As a good example, a multi-barrier treatment (MBT) comprising the steps of filtration, photolysis of hydrogen peroxide (H_2O_2/UVC) and catalytic wet peroxide oxidation (CWPO) using granular activated carbon (GAC) as catalyst demonstrate a remarkable efficiency for synthetic industrial effluents post-treatment, particularly because of the possibility of removing residual H_2O_2 and some by-products generated during H_2O_2/UVC process through subsequent filtration by GAC column [113,114]. Accordingly, Rueda-Márquez et al. [115] investigated the viability of a MBT process for post-treatment of plywood mill effluent on a pilot scale. As a result, over the MBT process, the concentration of COD and TOC decreased by 88.5% and 76%, respectively, while SS and turbidity decreased by 89% and 70%, respectively. Moreover, an almost complete H_2O_2 removal (95%) was achieved after CWPO process (1.5 min of contact with GAC). On the other hand, the operation and maintenance cost for this MBT process was estimated as 0.95 €/m^3

2.3.2. Bio-chemical combined treatment methods

Former studies have clearly indicated that the application of either a single biological or a single chemical method would not be enough to complete treatment of the P&P mill effluents in order to satisfy legal requirements and protect the environment [1]. Besides some species of single bacteria, mixed culture acclimated biomass as well as biofilms can partially or completely degrade adsorbable organic halides (AOX). Considering the fact that AOX has a severe toxic effect on the growth of microorganisms, combining two process; adsorption and biofilms, more satisfactory results can be achieved for AOX treatment. On the other hand, HRT is one of the most important parameters that should be considered for achieving optimal conditions for wastewater treatment. Osman et al. [116] by combining adsorption and biological processes at a lab-scale granular activated carbon sequencing batch biofilm reactor (GAC-SBBR) under aerobic conditions, tried to evaluate the effect of HRT on the GSC-SBBR performance in terms of the simultaneous removal of AOX and COD from a recycled paper mill wastewater. For this purpose, the reactor was packed with 80 pieces of cylindrically shaped plastic media with GAC of sizes in the range of 6–8 mm. These packing materials played two roles, both as adsorption medium and as a medium for biofilm growth. The results indicated that just a meagre efficiency can be achieved in the COD removal when HRT reduces from 48 h to 8 h. The results also demonstrated that the treatment efficiency for the reduction of COD and AOX from P&P mill effluents with GAC-SBBR can be obtained were as high as 92% and 99%, respectively, at an HRT of 48 h.

Addition of trace elements to the biological treatment systems has also been a solution to enhance the efficiency of the treatment, especially when facing with a highly polluted and complex effluent [117], as also previously observed for other types of industrial effluents [118,119]. This idea was tested by Barnett et al. [120] to promote the effluents from a TMP process by adding trace metals including Ca, Co, Cu, Fe(III), and Mg to an activated sludge (AS) plant as one of the common biological treatment to deal with P&P mill effluents [121]. They indicated that such trace elements are capable to enhance the COD removal from 82% to 86% (1.0 mg/L). Table 3 provides a summary of the recent studies on the application of non-additive methods for the treatment of pulp and paper mill effluents.

3. Challenges and opportunities

Currently, P&P mills continue to seek sustainable management solutions to overcome their difficulties in production processes and also to deal with economic and environmental management issues [124]. It is also of high importance for a good solution to establish a logic connection between the process engineering and economic considerations in line with the adoption of the best suited treatment process. Several techno-economic analysis have been carried out recently in order to optimize the production processes to have less environmental drawbacks while providing more quality for the products in an economic perspective [125–131]. But, industry is not yet able enough to minimize the pollution load in final effluents and it is expected for effluents from P&P industry to remain as one of the most polluted industrial wastewaters through the world containing recalcitrant and complex organic compounds [132]. There are a number of evidence for the soil pollution of the pulp and paper mills surrounding environment and severe toxic effects for the local biotic communities if discharging their wastewaters without efficient treatment [117,133–138]. In this situation,

Table 3
A summary of recent non-additive methods applied for the treatment of pulp and paper mill wastewater.

Method	Experimental conditions					Parameters									Ref.
	Effluents origin	Time	pH	Additive(s)	Temperature (°C)	COD		BOD		Colour		Other parameters			
						Initial (mg/L)	Removal (%)	Initial (mg/L)	Removal (%)	Initial (pt-Co)	Removal (%)	Parameter/ substance	Initial	Removal (%)	
UI&F	Raw Bleaching effluent	60 min	6.9	Fe(III): 1 g/L H_2O_2: 3 g/L		1510	12								[107]
PF							20					Abietic acid		93	
												Linoleic acid		84	
												β-sitosterol		87	
Electrochemical treatment							28					Abietic acid		51	
												Oleic acid		76	
												β-sitosterol		83	
Sono-electrocoagulation		4 h	7.5	Fe/Fe-electrodes		3000	95	1000	95	Dark brown	Color-less	TSS	2000 (mg/L)	87.5	[108]
												Odor	Burnt sugar	Nil	
Filtration fenton like oxidation	Fiber sewer collection unit effluent		3	H_2O_2 = 15 mM, Fe (II) = 6 mM		1943	95.02	250				Lignin		> 90	[109]
												UV_{254}		86.74	
												UV_{280}		87.08	
Filtration/$S_2O_8^{2-}$/Fe(II)			6	$S_2O_8^{2-}$ = 7 mM Fe (II) = 2 mM			94.96					Lignin		> 85	
												UV_{254}		92.04	
												UV_{280}		90.16	
UF and AOP	Acidic bleaching effluent		2.5	H_2O_2 BDD Anode	25	1250		563				TOC	499 (mg/L)	68	[41]
NF and AOP	Alkaline bleaching effluent		10.5	H_2O_2 BDD Anode	25	1500		595				TOC	594 (mg/L)	96	[41]
Electro-coagulation and UV/persulfate or UV/ peroxymonosulfate			8.2	Persulfate Peroxymonosulfate	23–27	1537	61	410							[110]
Filtration and AOPs	Plywood mill effluent	5 min		H_2O_2: 200 mg/L		230	88.5					SS	26 (mg/L)	89	[115]
												TOC	70 (mg/L)	76	
												Turbidity	8.3	70	
Adsorption and aerobic process (GAC-SBBR reactor)	Recycled paper mill (Dissolved air floatation)	48 h	7.1	Granular activated carbon		1152	92					AOX		99	[116]
Electrocoagulation		60 min	5	Iron & Aluminum	Room temperature	657	85			Brown	Completely	Lignin	1,155 (mg Phenol/L)	78.5	[122]
Electrocoagulation	Tissue paper wastewater		7.4	Iron, Aluminum and Sludge		1220	92.6								[123]

the end of pipe treatment of the wastewater will be considered, at least in the forthcoming years, as the main way to satisfy environmental protection communities. The stages involved in the treatment of such highly polluted and complex effluents are currently subjects of negotiations among the scientific communities. Some studies emphasizes that a series of biological treatments are good candidates to achieve the treatment goals regarding the stringent environmental protection standards and regulations [139]. For instance, Buyukkamaci and Koken in the year of 2010 [140] by comparing ninety-six treatment plants (in Turkey) having a single physical treatment, chemical treatment, aerobic and anaerobic biological processes or their combinations according to economic criteria including investment, operation, maintenance and rehabilitation costs tried to suggest an optimum treatment method for the P&P industry. They concluded that an extended aeration activated sludge process is the best option for low strength effluents, while extended aeration activated sludge process or UASB followed by an aeration basin can act efficiently for medium strength effluents, and finally UASB followed by an aeration basin or UASB followed by a conventional activated sludge process can optimize the techno-economic considerations for the treatment of high strength P&P effluents. However, the necessity of allocating a relatively large area for biological treatments, relatively long-time treatment requirements as well as uncompleted treatment and local issues such as the bad smell resulting from the bacterial activities are the main drawbacks of such systems. In addition, conventional biological treatments have shown a limited efficiency for the treatment of recalcitrant and complex pollutants such as AOXs which can remain in the treated effluents, causing several environmental and health problems [141,142]. Hence, there is also a need to perform further life cycle assessment studies for the biological treatment methods applied to this type of effluents [143,144]. The scientific orientation observed by the researchers in very recent literature emphasizes that the emerging novel technologies could considerably eclipse the idea of using sequential biological treatments. Economic analysis performed in some physico-chemical treatments methods, such as Fenton process [145,146], indicated their cost-effectiveness. In addition, with respect to adsorption processes it has been reported in the literature that activated carbons, among the existing adsorbents, can offer a high adsorption performance due to its specific characteristics such as large surface area and pore volume, high-speed kinetics and coarse texture [9,10]. However, although in comparison with relatively expensive adsorbents such as silica, activated carbon can contribute to minimize the overall treatment cost, it may be here suggested the need to develop the application of cheap materials that are by-products of other natural or artificial chemical-based processes as adsorbents. Hence, it is also recommend wider applications of biochar not only because of its low cost and high performance but also due to its potential to reduce toxicity to the environment (such as soil) [14] after the treatment process. In this regard, ashes can also be a good candidate to this end, containing relatively high amounts of metallic elements such as Fe, Ca, Na, etc. Besides their ability to adsorb contaminants from the polluted effluents, they readily release relevant cations to the media. This can promote the treatment efficiency via inducing chemical reactions with the release of electrons from the metallic elements in the media such as those (such as iron ions) used for the removal of toxic materials from the effluents such as nitrate [147–149]. They can also provide enough nutrients in the final sludge content, making them suitable for land-use utilizations, for instance for agricultural applications which may need a sludge with a minimum content of toxic substances and enough amounts of nutrient elements [150,151]. The wastes from wastewaters treatment plants (such as secondary sludge) can also be considered as a low cost solution to adsorb recalcitrant compounds. However, the final sludge after the adsorption process can be harmful to the environment due to the attachment of toxic compounds to its surface. In this case, probably the incineration is the only existing way to deal with the final wastes, which will cause some environmental drawbacks [152]. In the other hand, some novel technologies such as the application of mesoporous nanomaterials with high adsorption capacity [152–154] can also make a revolution in the treatment of highly polluted and complex effluents from pulp and paper industry. In this regard, enough engineering efforts must be done to maximize the efficiency of such materials for better treatment results. This can be achieved by adopting optimization techniques such as surface response methodology [152–154] to reach best results in a cost-effective manner. Such efforts can also be made for the synthesis of materials with high optical properties to reduce the need for irradiation of light to the reaction medium during the processes such as oxidation of effluents. In this way, economic considerations of the treatment process can be also satisfied.

The combination of various additives can also result in the promotion of the treatment efficiency. Also, the combination of various additive-based methods with biological methods have shown attractive results for the elimination of recalcitrant pollutants from P&P mill effluents such as AOXs and to promote the overall efficiency of COD and BOD removals, which can result in a better quality for the final effluents. There is also a need to design combined reactors to join physico-chemical and biological methods by taking into consideration that such systems must not only enhance the overall treatment performance, but also be technically and economically feasible enough to encourage investors. This latter item is currently the main obstacle for the rapid commercialization of the lab-scale developed novel methods, especially in developing countries.

Application of chemical additives can generally raise some concerns on their toxic effects when released to environment [155]. Furthermore, the review of recent studies on the treatment of P&P mill effluents reveals a trend for the use of chemical additives but without enough eco-toxicological studies. This issue is of high importance because recently the concern on the effects of the remaining toxic chemicals after the treatment process has increased considerably in the scientific community. Hence, there is an urgent need to carry out life cycle assessment studies for emerging additive-based technologies for the treatment of highly polluted and complex industrial effluents such as those released to environment from the P&P industry.

It is also worthy to mention that the selection of a sustainable method to deal with effluents from pulp and paper industry is a complex task, requiring the consideration of various technical, environmental and economic as well as social criteria. Hence, application of multi-criteria decision making systems (MCDM) [156,157] is highly recommended in this regard to find the best available technologies for the treatment of pulp and paper mill effluents.

4. Conclusion

This paper presents very recent results achieved on the treatment of pulp and paper mill effluents based on the application of chemical additives alone or in combination with other physical or biological treatment methods. Moreover, recent developments on single biological and physical treatments or their hybrid application have been critically reviewed. As a result the interest of the scientific community to promote additive-based treatment methods in combination with conventional or novel physical or biological treatment methods could be highlighted. The overall trend in this field, as realized by the authors, is to promote techno-economic benefits of treatment processes. Furthermore, taking into account toxicology and environmental safety issues this review calls for joint studies or complementary studies on the subsequent environmental impacts of the novel combined technologies towards an efficient treatment method.

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