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Comparison of different approaches for color and COD removal from paper and pulp industry effluent

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ABSTRACT

Effluent from the paper and pulp industry has been treated by various single and integrated approaches. Single approaches include sand filtration, algal treatment, chemical coagulation, electrocoagulation, and ozonation. Whereas the combinations of pre-chemical coagulation and sand filtration, pre-chemical coagulation and ozonation, post-chemical coagulation and ozonation, and pre-electrocoagulation and sand filtration were included in integrated approaches for determination of COD and color removal efficiencies. The application of sand filtration, algal treatment, chemical coagulation, electrocoagulation and ozonation have been deliberated the removal efficiencies (%) for COD as 8.73, 78.18, 16.96, 70.37 and 81.19 and for color as 9.09, 48.72, 58.57, –53.48 and 81 respectively. Subsequently the applications of combinations of pre-chemical coagulation and sand filtration, pre-chemical coagulation and ozonation, post-chemical coagulation and ozonation, and pre-electrocoagulation and sand filtration have resulted in the removal efficiencies (%) for COD as 97.33, 56.12, 88.04 and 70.33. Color removal efficiencies were 76.36, 76.36, 78.18 and 67.15 respectively. The integration of alum coagulation and sand filtration were found to be most effective and economical methods.

Keywords: Integrated techniques for wastewater treatment; Paper and pulp industry; Ozonation; Coagulation; Sand filtration; Algal treatment

1. Introduction

The pulp and paper manufacturing industry is a high water consuming chemical industry and rated third after metal and chemical industry based on its water consumption pattern [1,2]. All aspects of environment such as land, air and water are affected by the effluent discharged from the industry. Historically paper and pulp industry has been thought to be a chief consumer of energy, natural resource and a major contributor of pollutant released to environment [3]. Paper and pulp industry is second largest industry in Pakistan after textile. This industry consumes excessive amount of water approximately 60 m^3 per ton of paper produced. Wastewater treatment facilities are inadequate in almost paper and pulp industry in Pakistan [4].

Effluent discharged from pulp and paper industry contains a high amount of pollutant such as COD, BOD, color, chlorinated compounds, sulfur and sulfur compounds, suspended solids mainly fatty acids, fibers lignin and its derivatives. Out of these described pollutants, COD, toxicity, suspended solids and color cause major environmental problems [5,6]. High molecular weight of synthetic dyes and bleaching agents are responsible for huge amount of refectory COD. Level of toxicity, total organic carbon (TOC), COD and color removal by conventional treatment vary widely depending on the pulping method used [7,8]. A novel approach should be developed for the wastewater treatment to deal with strict environmental regulations on effluent quality entering recipient water bodies [9,10].

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Vigorous wastewater treatment approaches need to be implemented prior to the final dumping of wastewater into natural water bodies. The treated effluent should meet the water quality standards in order to reuse and reduce fresh water intake [11]. Conventional biological treatment methods are not adequate for treatment, as some of the pollutants in pulp and paper industry effluent are non-biodegradable. Removal of color in effluent is not possible with conventional biological treatment in some cases it is observed that these treatments increase the color of effluent wastewater [12].

Combined treatment strategy can have better potential to treat less biodegradable effluent over single-step treatment method alone. By using integrated approaches we can take advantages of unique characteristics of more than 1 treatment method [13]. Combination of biological treatment, coagulation with alum and sand filtration generate good results in pilot scale reactor. Integration of chemical precipitation followed by ozonation also proved to be effective [14]. Integration of aerobic treatment followed by anaerobic treatment can reduce color and TOC [15]. Integration of physio chemical treatment processes removes toxic pollutants. A long-term solution for paper and pulp industry is to combine biological and physio chemical treatment processes [16]. Jaafarzadeh et al. [17] also resulted that integration of coagulation and electrochemical process can upsurge the bio-degradability wastewater of paper and pulp.

The objective of the present study was to compare COD and color removal efficiencies of single and integrated approaches from effluent of pulp and paper industry.

2. Materials and methods

In order to carry out the experiments the wastewater samples were collected from the Century pulp and paper industry located near Lahore, Pakistan. Effluent samples were collected from industry by using grab sampling technique. Samples were collected in December. Random samples were taken from the outlet of dissolved air floatation plant in dry and sterilized plastic containers. Samples were stored in refrigerator at 4°C in Wastewater Laboratory at Sustainable Development Study Center, Government College University, Lahore. Initial amount of COD and color were measured in the laboratory.

2.1. Determination of lambda max

To determine the color of effluent lambda max was determined. Lambda max is the wavelength at which the maximum fraction of light is absorbed by a solution. The lambda max was determined using Spectrophotometer (Spectroscan Company, 80D model), colorimetric method. At different wavelength, absorbance of sample was measured. The wavelength (420 nm) with maximum absorbance (0.7 with 5 times dilution) was considered to be the lambda max. Then the absorbance of the different treated samples were measured and compared with lambda max.

Chemical Oxygen Demand was determined by COD reactor (Model: Lovibond, ET108), method 5220 according to standard method of APHA [18].

2.2. Experiments

Experiments were carried out using different single and integrated wastewater treatment approaches. Five different single treatment options were used separately. In integrated treatment methods, different combinations of single treatments were used to check the percentage removal efficiencies of color and COD.

2.3. Single treatment methods

2.3.1. Sand filtration

In this typical method, a gravel and sand filled column of 3 ft height and (1.181 inch) was used. The column was filled with gravel and sand up to 2.5 ft. The sand was passed through sieves of 100 µm and 200 µm pore size. Water sample was passed through the clean sand and gravel layers, and after filtration the COD and color removal efficiencies were determined.

2.3.2. Algal treatment

In algal treatment, different ratios of industrial effluent and sewage water (1:0, 4:1, 3:2, and 2:3) were used and 20 g algae were added in each container to grow for one week. It was a mixed culture of algae dominated by *Chaetomorpha Sutoria*, *Sirogonium Sticticum* and *Zygnema Sp* which were collected from GC University Lahore's Botanic Garden. Yasar et al. [19] also used these species for removal of pollutants in wastewater. The effluent was treated with this mix culture in combination with sewage water in 1000 ml plastic containers. Samples were taken on daily basis to check the efficiencies of COD and color removal.

2.3.3. Chemical coagulation

Two Pyrex glass beakers of 1000 mL were used. 1000 mL sample was taken in each beaker and alum dosage of 2 g/L and 4 g/L were introduced into these beakers. Different alum dosages were added into the sample and stirred at different rates. Samples were stirred for 1 min at 100 rpm, again beaker were stirred for 10 min at 80 rpm and were allowed to rest for 1 h. After settling of the coagulants, the samples were taken to observe the color and COD removal efficiencies.

2.3.4. Electrocoagulation

Electrocoagulation was carried out by using different arrangements of iron and aluminum electrodes. NaCl was added as electrolyte. Water samples were stirred, filtered and rested for sedimentation. After that samples were analyzed for COD and color removal. Electrolysis time and electrodes selection were changing parameters but distance between electrodes was kept constant. Small variations in current intensity and voltage were observed. Mechanism of electro coagulation is summarized as follows:

$$Fe_s \rightarrow Fe^{3}_{aq} + 3e^- \text{ (for iron electrode)} \quad (1)$$

$$Al_s \rightarrow Al^{3+}{}_{aq} + 3e^- \text{ (for aluminum electrode)} \tag{2}$$

The reaction occurs among ionic species to produce flocks i.e. Al $(OH)_3$ and Fe $(OH)_2$

$$Fe^3{}_{aq} + 2OH^- \rightarrow Fe(OH)_{3s} \text{ (for iron electrode)} \tag{3}$$

$$\begin{matrix} Al^{3+}{}_{aq} + 3H_2O \rightarrow \\ Al(OH)_{3s} + 3H^+{}_{aq} \end{matrix} \text{ (for aluminum electrode)} \tag{4}$$

2.3.5. Ozonation

As strong oxidizing agent ozone has such an oxidation mechanism, in which ozone reacts directly with dissolved compounds or with radicles produced depending on the pH of the solution [20]. Ozone was generated by ozone generator of model OZ. Ozone column reactor was of 3 cm diameter and 132 cm height. 500 ml of filtered and unfiltered samples were added to ozone column reactor in the process of ozonation. Samples were treated at pH 5, 6.7 and 9. To estimate the color and COD removal efficiencies, samples were taken from the ozone column reactor after regular time intervals.

2.4. Integrated treatment methods

2.4.1. Pre-chemical coagulation and sand filtration

Pre-chemical coagulation and sand filtration methods had been integrated in such a way the sample was firstly treated by using the process of chemical coagulation and secondly the effluent from this process was filtered through the sand and gravel column. 250 ml of sample was taken in a beaker and optimized dose of alum (0.5 g) was added in it. A column of 3 ft height and (1.181 inch) diameter was used for sand filtration. The column was filled with gravel and sand up to 2.5 ft.

2.4.2. Pre-electrocoagulation integrated with sand filtration

In the combination of pre-electrocoagulation and sand filtration the sample was passed through the electrocoagulation process where cathode and anode of aluminum were used. Treated sample was then passed through the gravel and sand filled column. Filtered sample from both integrations, were tested for COD and color removal efficiency.

2.4.3. Pre-chemical coagulation integrated with ozonation

This typical mechanism employed the sample preparation by means of stirring and settlement under the process of chemical coagulation. After that the same sample was used for ozone treatment for few minutes. At the end of both processes, sample was withdrawn to measure color and COD removal efficiency.

2.4.4. Post-chemical coagulation and ozonation

In combination of post-chemical coagulation and ozonation, the sample was first taken for ozone treatment then chemical coagulation process was applied. After both of the applications, the sample was collected to check for color and COD removal efficiency. 500 mL of sample was taken for ozone treatment at its normal pH (6.7) for 5 min, as these were the optimum reaction conditions.

3. Results and discussion

Table 1 shows the effluent characteristics of raw water from paper and pulp industry with exceeding limits of COD as compare to Punjab Environment Quality Standards 2016. The research was mainly focused to remove color and COD of the pulp and paper industry wastewater by using different single and integrated methods at low cost and energy, so to determine the most economical and efficient color and COD removal approach.

Fig. 1 shows a comparison of optimum removal effectiveness and relative efficiencies obtained by all single approaches.

3.1. Analytics of single treatment methods

3.1.1. Effectiveness of sand filtration

While treating the effluent of pulp and paper industry wastewater with sand filtration mechanism the resultant efficiencies of COD and color (8.73% and 9.09%) were not found significant. The wastewater moves faster through the sand due to the large size grains of sand. Small medium is time consuming and chances of clogging are higher because of slow water movement. Solid particle penetration and quality of final effluent is dependent on grain size [21].

3.1.2. Effectiveness of algal treatment

In algal treatment method, different effluent to sewage ratios were used to check the removal efficiencies of

Table 1
Effluent characteristics from paper and pulp industry

Sr. #	Parameter	Values	*PEQ's 2016
1	Temperature, °C	22 ± 2	≤30
2	Salinity, ppt	1.8	ND**
3	Electrical conductivity, µS	2.7	ND**
4	Dissolved oxygen, ppm	0.00	ND**
5	pH	6.7	6–9
6	Turbidity, FTU	142	ND**
7	Total settle able solids, mg/l	0.5	ND**
8	Total dissolved solids, mg/l	200	3500
9	Total suspended solids, mg/l	133	200
10	Chemical oxygen demand, mg/l	2393	150
11	Biological oxygen demand, mg/l	1628	80
12	SO_4^{-2}, ppm	368.8	600
13	PO_4^{-2}, ppm	1.43	ND**

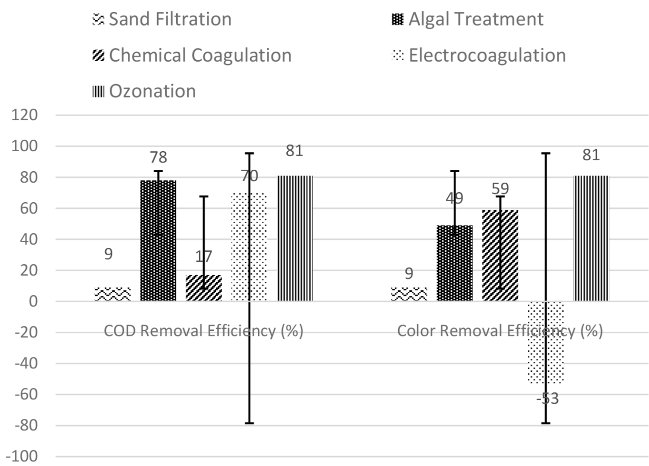

Fig. 1. Comparison of removal efficiencies of COD and color for different single treatment methods.

COD and color. Maximum removal of COD and color was in 4:1 and 2:3 samples (78.18% and 21.21%, 70.83% and 48.72%) respectively. Table 2 shows color and COD removal efficiencies at different effluent to sewage ratios. Wastewater treatment through algae has been in exercise over forty years. Comparison can be made among other conventional technologies and microalgae on the basis of transformation removal and degradation of toxic and non-toxic pollutant [22]. The benefit of algal treatment is that it is an aerobic process and unlike the anaerobic process, it treats the wastewater rapidly without giving unpleasant odor [23,24].

3.1.3. Effectiveness of chemical coagulation

Different dosages of alum were used in the process of chemical coagulation. Sample of wastewater was treated with 0.5 g alum dose and the removal efficiencies of COD and color were 16.96 and 58.57 respectively. By doubling the alum dose removal efficiencies were decreased as shown in Table 3. For treatment of wastewater most common coagulants used were salts of iron and aluminum [25]. Color and COD removal efficiencies increased with increase in alum dosage until they reached at optimum dose and after that removal of pollutants decreased by increasing the alum dose. Coagulation with alum is effective for color removal from pH 4 to pH 12. Application of alum dose showed similar trends for COD and color, which confirms that organic matter, is responsible for color production [26,11]. Commercial alum is the most effective coagulant at 5 kg/m^3 of dosage [27].

3.1.4. Effectiveness of electrocoagulation

In electrocoagulation different combinations of electrodes were used to check the effect of treatment on removal efficiency, at different time intervals, with constant concentration of NaCl electrolyte. Highest removal efficiencies (%) were achieved 70 for COD and 53 for color, when both electrodes were of aluminum and the time was 6 min. Table 4 shows the results of COD and color removal efficiencies

Table 2
COD and color removal efficiencies at different effluent to sewage ratios in algal treatment method

Effluent : Sewage	% Removal efficiency	
	COD	Color
1:0	69	0
4:1	78	21
3:2	59	47
2:3	71	49

Table 3
COD and color removal efficiencies at different alum doses in chemical coagulation method

Alum dose (g)	% COD removal efficiency	% Color removal efficiency
0.5	17	59
1	11	54

when different combinations of electrodes were used at different time.

Iron oxidizes immediately and brings dark color to the sample being treated thus intensity of color increased instead of decreasing. Maximum COD removal efficiency was achieved at pH 7 and beyond that pH solubility of $Fe(OH)_3$ increased resulting in formation of soluble $Fe(OH)_4$ which did not contribute to the COD and color reduction [28]. Efficiency of electrode is dependent on electrode type, reaction conditions, setup for treatment, and its integration with other treatment methods like advanced and enzymatic oxidation processes [29].

3.1.5. Effectiveness of ozonation

In the treatment process by ozonation the removal efficiency was determined by varying pH of effluents where the concertation of ozone was constant. Maximum COD reduction was observed at pH 9 which was 81% and maximum color removal efficiency was 81% at pH 6.7. In Table 5 result of COD and color removal efficiencies at different pH and are presented. The concentration of ozone was kept constant throughout the experiment. Removal efficiencies increased with increase in pH of the solution. In alkaline conditions ozone converted to hydro peroxide anions which resulted in higher removal efficiencies. OH· radical has high oxidation potential which is very important in the decolorization process [30]. Generally ozone oxidation pathways consist of a direct oxidation by ozone or a radical oxidation by OH· radical [31]. Major limitation of ozonation treatment method is its high cost and short half-life. Due to the low solubility of ozone in aqueous solutions it may be difficult to achieve efficient gas liquid mass transfer [32]. Color and COD removal trend were different for all single treatments.

Fig. 2 represents the comparison of COD and color removal efficiencies from different integrated treatment methods.

Table 4
COD and color removal efficiencies at different time with different types of electrodes by applying electrocoagulation at 12V

Type of electrodes	Reaction time (min)					
	2		4		6	
	% Removal efficiency					
	COD	Color	COD	Color	COD	Color
Fe plates	60	–111	67	–111	69	–111
Al plates	69	–88	70	–70	70	–53
Fe anode and Al cathode	44	–178	51	–178	58	–178
Fe cathode and Al anode	47	–111	54	–74	71	71

Table 5
COD and color removal efficiencies at different time and pH by applying ozonation treatment

Time (min)	pH					
	5		6.7		9	
	% Removal efficiency					
	COD	Color	COD	Color	COD	Color
5	62	74	76	76	80	80
10	67	76	77	80	81	81
15	72	79	79	81	81	81
20	78	79	79	81	81	81

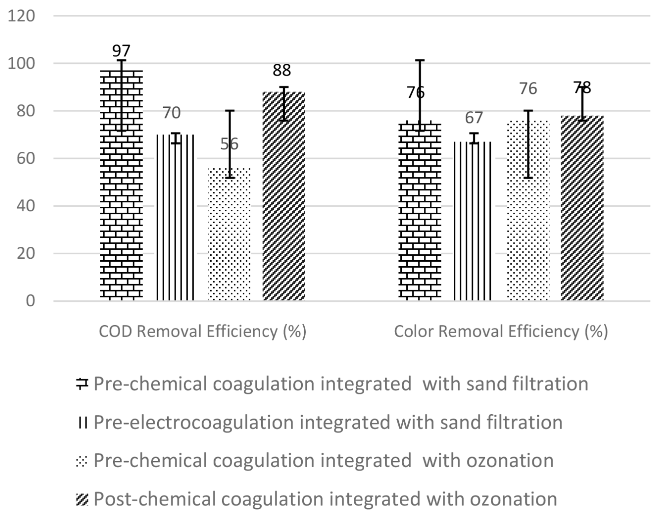

Fig. 2. Comparison of removal efficiencies of COD and color for different integrated treatment methods

3.2. Analytics of integrated treatment methods

3.2.1. Efficiency of pre-chemical coagulation and sand filtration

In integrated approach of pre-chemical coagulation and sand filtration, optimum dose of alum (0.5 g) was used for coagulation. Removal efficiencies (%) for COD and color were 97 and 76 respectively. Formation of large colloidal particles in the coagulation process can be responsible for color and COD removal. Presence of suspended particles responsible for COD removal in the treated effluent can be further removed by slow sand filtration.

3.2.2. Efficiency of pre-electrocoagulation and sand filtration

In combination of pre-electrocoagulation and sand filtration, electrocoagulation was carried out by using both aluminum electrodes. The supernatant from this process was further treated through the filtration column. Removal efficiency from this process was 70% for COD and 67% for color. Electrochemical degradation as compared to traditional coagulation techniques has ability of removing smallest colloidal particles. Sand filtration further removed the color which increased due to the oxidized electrodes [33].

3.2.3. Efficiency of pre-chemical coagulation integrated with ozonation

In integrated treatment approach of pre-chemical coagulation and ozonation, optimum dose of alum (0.5 g) was used for coagulation. The sample obtained from this process was further treated in ozonation column at 6.70 pH. Calculated removal efficiencies were 56% and 76% for COD and color respectively. Alum reduces the pH of the effluent and makes it acidic. Ozonation is more effective treatment approach for the effluent with higher pH [34].

3.2.4. *Efficiency of post-chemical coagulation integrated with ozonation*

In the combination of post-chemical coagulation and ozonation, the wastewater was treated in ozonation column for 5 min at 6.70 pH. The ozonated sample was further treated for chemical coagulation by using optimum dose of alum (0.5 g). The procedure gave 88% COD removal efficiency and 78% color removal efficiency. Application of pre-ozonation reduces the amount of coagulant required for treatment which decreases colloidal charge density during oxidation. Ozonation may oxidize the organic substances of high molecular weight and convert them to smaller one [35].

4. Conclusion

The relative analytics and results revealed that effective methods in single treatments were; algae > ozonation > electrocoagulation > coagulation. However the percentage removal efficiency was found to be maximum in ozonation (color removal 81.19% and COD removal 80.90% at 20 min, pH 9) but this technique is relatively expensive as compared to algal treatment. Algae utilization was found to be the cost effective option as it removed the maximum color and COD in 4:1 (78.18% and 21.21%) and 2:3 (70.83% and 48.72%). Whereas in integrated techniques, the maximum color removal efficiency (78.18%) was obtained in pre-chemical coagulation and ozonation, even though the maximum COD removal efficiency (97.33%) was observed in-case of sand filtration and pre-chemical coagulation technique. The relative effectiveness for this integrated treatment has been determined in the order of; pre-chemical coagulation and ozonation > sand filtration and pre-chemical coagulation > pre-electrocoagulation with sand filtration > pre-chemical coagulation with ozonation.

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