Combined Biological and Advanced Oxidation Treatment Processes for COD and Color Removal of Sewage Water

C. T. Mehmood, A. Batool, and I. A. Qazi

Abstract—Wastewater (WW) generation is inevitable in rapidly growing and urbanizing societies resulting in significant damage to the quality and quantity of fresh water resources. The study was intended to harness the reuse potential of sewage WW using integrated wastewater treatment process. Composite sample of sewage WW was collected from inlet of sewage wastewater treatment plant. The WW sample was batch wise subjected to activated sludge process (ASP), ozonation, UV irradiation, H_2O_2 oxidation and sand filtration separately for different time intervals (i.e. 0 to 60 min) and concentrations of H_2O_2 (0.1 to 1.0 mL^{-1}), followed by the integration of all the processes at their optimum conditions. Batch experiments were performed in pyrex containers using 1.5L sample except for ASP that is 20L. Efficiencies were measured in terms of COD, color and TSS removal along with changes in pH and temperature. Biological diversity of WW was also determined using cultural techniques followed by biochemical tests. Color reduction was much higher and faster than COD in all the tested treatment processes. Ozonation was the fastest process to remove 78% color in 30 minutes however, ASP was more economical for the same efficiency but requires high hydraulic retention time (HRT). Increase in UV irradiation time reduced COD and color by 54 and 69% after 60min respectively. Sand filter efficiently removed 97% TSS with marginal reduction in COD. Integration of 4 hr ASP, 24 min ozonation and 10 min UV exposure with 1 mL L^{-1} H_2O_2 dose yielded 98 and 100% COD and Color reduction respectively.

Index Terms—Advanced oxidation, COD, color, sewage water.

I. INTRODUCTION

Wastewater generation is inevitable in rapidly urbanizing areas like Pakistan having present population of 155 million which is expected to be 229 and 295 millions in year 2025 and 2050 respectively. Estimates suggest that Pakistan had about 5,000 m^3 per capita per annum of fresh water in 1951, 1,500 m^3 per capita per annum at the moment, and expected to drop less than 1,000 m^3 per capita in 2020 to be declared as water stressed country [1]. Furthermore, access to safe drinking water and sanitation facilities are available to only about 65% and 59% of total population respectively [2]. It

has been estimated that around 2000 million gallons of sewage is being discharged to surface water bodies daily, while collection level is estimated to be no greater than 50 % nationally (less than 20 % in rural areas), however, only 10 % of collected sewage is effectively being treated [3].

Treatment of wastewater is pricy and difficult due to quality and composition [4]. Many different traditional treatment techniques are being applied on wastewaters such as coagulation/flocculation, membrane separation, ultra filtration, reverse osmosis, microfiltration, activate carbon adsorption and sand filters [4]. However, none of these treatment methods are effective enough to produce water with acceptable levels of the most persistent pollutants (e.g., phenols, pesticides, solvents, household chemicals and drugs, etc.) [5]. Elimination of these compounds by activated carbon produce secondary pollutants, whereas biological treatment is widely used method for removing organic pollutants and some nutrients (N and P) from domestic effluents however, it is not a complete solution to the problem due to biological resistance of some dyes and inorganic chemicals [4]. Biological methods especially advanced nutrient removal systems are very complicated and difficult to stabilize and optimize especially when a toxic component is involved in the influent. In addition, these processes require a larger construction area and longer time for treatment. Furthermore, disposal of excess biomass is a significant cost component and conventional biological treatments often prove inadequate for complete removal of the pollutants [6].

A further treatment stage is mostly necessary to attain ample removal efficiency. Application of advanced oxidation processes (AOPs) can better serve the purpose as AOPs are recommended when wastewater components have a high chemical stability and/or low biodegradability. A chemical wastewater treatment using AOPs can produce the complete mineralization of pollutants to CO_2 , water, and inorganic compounds, or at least their transformation into more innocuous products [7].

All the advanced oxidation processes (AOPs) involve the generation of hydroxyl radicals [8]. These radicals are very reactive, attack most organic molecules, and are not highly selective [6]. Hence, the advanced oxidation processes (AOPs) like $\text{H}_2\text{O}_2/\text{UV}$, Fenton, photo-Fenton and photo-catalytic processes could be preferred options to treat and eliminate toxic and hazardous compounds from wastewater. The processes have the capacity to decolorize completely and partially mineralize the wastewater in short reaction time [9]. Fenton and photo-Fenton reactions are fast and efficient; however, they only work in acidic solutions and need extraneous iron (which has to be subsequently

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removed). Photo-catalytic processes also have efficient dye decoloration/degradation capability but the main drawback is the need for a relatively expensive TiO_2 catalyst and its subsequent removal [10].

Hydrogen peroxide with UV light has been found to be effective in treating mixed wastewater as it reduces COD, BOD and toxicity of mixed wastewater [11], [12]. Like other AOPs, H_2O_2 /UV process generates hydroxyl radicals and these $\text{OH}^\bullet$ radicals (due to their high oxidation potential) change nature of organic molecules either by removing H^+ or by adding it to double bonds present in toxic molecules [13]. The photolysis of hydrogen peroxide is preferred over other AOPs due to some advantages such as the complete miscibility of H_2O_2 with water, the stability and commercial availability of hydrogen peroxide, no phase transfer problems and lower investment costs [14]-[16]. UV light accelerates the rate of decomposition of H_2O_2 with corresponding increase in the rate of $\text{OH}^\bullet$ radical formation [12]. Additional advantage is that no residual products are formed and the treatment can be carried out under ambient temperature and the oxygen formed during the process is useful for aerobic biological treatment [6].

The objectives of the study were to investigate the effectiveness of individual WW treatment processes in local conditions and to estimate the effectiveness of integrated wastewater treatment process for improving the wastewater quality for reuse.

II. MATERIALS AND METHODS

A. Sample Characteristics

Composite wastewater samples were collected from inlet of sewage treatment plant (STP), I-9 Islamabad. Temperatures of the wastewater samples were measured on site and stored at 4 C. Wastewater was characterized using standard methods (Table 1). Biological diversity of WW was determined using cultural techniques followed by biochemical tests.

B. Treatment Processes

The WW samples were batch wise subjected to activated sludge process (ASP), ozonation, UV irradiation, H_2O_2 oxidation and sand filtration separately for different time intervals and concentrations of H_2O_2 , followed by the integration of all the processes at their optimum conditions. Batch experiments were performed in pyrex containers using 1.5L sample except for ASP, for which 20L sample was used.

Process efficiencies were measured in terms of COD, color and TSS removal along with changes in pH and temperature. Kinetic degradation of the dissolved organic matter was determined using global parameters like color and COD. Rate constants of decoloration and removal of COD were determined using following exponential relation

$$\ln C/C_0 = -k t$$

where C was concentration of color or COD at time t , C_0 was initial concentration of color or COD, and t time [14].

C. Statistical Analysis

The data was analyzed using factorial design and treatment

means were compared by LSD using SPSS for Windows version 13.0.

TABLE I: PHYSIO-CHEMICAL CHARACTERISTICS OF WASTEWATER

Parameter	Units	Values	WHO Standard
COD	(mgL^{-1})	387	150
BOD	(mgL^{-1})	250	100
TSS	(mgL^{-1})	323	<100
TDS	(mgL^{-1})	793	500
EC	(μS)	1353	1000
Temperature	($^\circ\text{C}$)	30	
Hardness	(mgL^{-1})	453	500
DO	(mgL^{-1})	0.23	2.0
pH		7.53	6-8
Alkalinity	(mgL^{-1})	520	400

III. RESULTS AND DISCUSSION

A. Biological Process

Activated sludge process was conducted in a plastic container having maximum capacity of 25 liters. Air was supplied using air pump with two outlets via sand stone diffuser placed at the bottom of the reactor. Sludge was prepared before the treatment of actual wastewater sample. Samples were treated for the Hydraulic Retention Time (HRT) of 2, 4, 6, 8 and 10 hours on original pH and temperature and were not adjusted throughout the treatment process.

Color was reduced faster than COD achieving maximum 73 and 63 % respectively in 10 hrs (Fig. 1). During initial 4 hours of the treatment, color was reduced rapidly (slope $y=14.2$) followed by slower rate which is explained by slope $y= 2.9$. Similarly COD yielded slope $y=8.78$ in the first 6 hours and the rate was reduced by 77.4 % (i.e. $y=1.98$) whereas, strong correlation (0.887) was observed between COD and Color.

High removal efficiencies might be due to the utilization of some readily biodegradable compounds in wastewater by the aerobic microorganism in ASP reactor [17]. COD removal may also be influenced by air stripping caused by the air supply at the bottom of the reactor through spargers [18]. Decrease in color removal rate at HRT above 4 hrs might be due to the formation of colored organic byproducts during the treatment [19].

Fig. 1. Effect of HRT on Color and COD removal in ASP

B. Sand Filter

Traditional sand filter was prepared using coarse gravel, fine gravel and sand in plastic container having capacity of 5.5 liters. Filling materials (Gravel and sand) were washed with detergents followed by distilled water and air dried. COD, color and TSS removals were statistically significant at 95 % of confidence interval. Over 95 % TSS was removed with 68 % color and 26 % COD reduction (Fig. 2). Removal of TSS with the application of sand filter is basically due to the fine sand particles strongly packed to each other which do not allow the suspended solids to pass through. Color and COD removal is probably due to the interaction of biosand particles and bacterial activity [20].

C. UV Irradiation

Ultra violet treatment was carried out as batch experiment at ambient temperature in Photo-reactor comprising of Pyrex cylinder with internal diameter of 10 cm, height of 25 cm and a total liquid volume of 1.75 L. Ultraviolet lamp had nominal power of 8 W ($\lambda=254$ nm). The UV lamp was positioned in the center of cylinder as a light source.

Analysis of variance (ANOVA) for color and COD with the interaction of varying UV exposure time showed significant decrease in color and COD with increase in UV irradiation time. Maximum color and COD reduction was 76 and 61 % respectively after 120 min of UV irradiation (Fig. 3). Initially color and COD removal rates were faster i.e. 50 % of total color and COD reduction was achieved in just 20 min and 30 min respectively. These are depicted by slopes i.e. $y = 1.52$ and 1.09 for color and COD respectively. However, 94 % and 86 % decrease in the rates of color and COD were observed respectively after 50 min. Both color and COD were strongly correlated to UV irradiation time as well as to each other ($r=0.851$).

The high intensity of the UV light can change the physicochemical nature of the organic matter in the wastewater. The reduction in the COD and color can be the result of continuous exposure of UV light which provided the energy to the atoms resulting in wide range of chemical reaction. Compounds that absorb UV radiation within the corresponding range of the spectrum may be photo-degraded.

The costs of UV radiation treatment processes depend to a great extent on the absorption properties of the compounds to be eliminated [21]. Real wastewater generally has wide range of chemicals which have the light absorbing potential and are best suited for UV treatment. However, the application of UV light alone is not appropriate to reduce the contaminant contents of wastewater [22].

D. Ozonation

Ozonation was carried out in a reactor having similar specifications used for UV experiments. A lab scale ozonator with ozone output of 200-300 mg/h was used as ozone source. Maximum COD and color reduction was 41.7 and 76 % respectively after 30 min of ozonation (Fig. 4). Color removal slope ($y=2.22x$) was double as compared to the COD removal slope ($y=1.09x$). Ozonation process produces very reactive atomic oxygen which rigorously attaches the organics in the wastewater which ultimately results in the reduction of COD and Color. Further, ozone concentration

increases in the air bubbles with increasing time which serves as the driving force for the transfer of ozone to the wastewater with a consequent increase in ozone concentration in the solution and the rate of oxidation [23]. The decolorization of wastewater with increasing time is attributed to the enhancement of mass transfer of ozone from air-ozone bubbles to the liquid phase as a result of the ability of the rising bubbles to induce radial momentum [24] and increase in the gas hold up [25].

Fig. 2. Reduction in COD, color and TSS of wastewater using sand filter

Fig. 3. COD and Color removal of wastewater with increase in UV time

Fig. 4. COD and Color reduction with increase in ozonation time

Ozonation process was observed as potential option for wastewater treatment [26]. Faster decoloration than the COD reduction during the whole process may be due to the fragmentation of the initial structures of the aromatic colored compounds [3], [6].

E. Hydrogen Peroxide

Hydrogen peroxide dose was optimized in the pyrex container as described in UV experiment. Stirring was achieved by a rotor placed at the bottom of the reactor. Three levels of H_2O_2 (i.e. 0.1, 0.5 and 1.0 ml/L) were applied. Hydrogen peroxide application significantly decreased COD and color with increase in H_2O_2 concentration from 0.1 to 1.0 ml/L. Maximum COD and color reduction was 36 and 65 % respectively (Fig. 5). Chemical oxygen demand reduction was more correlated with the hydrogen peroxide dose (i.e. $R^2=0.926$) than the color removal ($R^2=0.675$). Hydrogen peroxide concentration plays a crucial role in deciding the overall efficiency of the degradation process.

It was observed that the percentage degradation of the pollutants increased with an increase in the dose of hydrogen peroxide [27]. The degradation of wastewater may be due to the generation of hydroxyl radicals capable of oxidizing the organic compounds present in the wastewater. Higher doses of H_2O_2 produce more number of OH radicals which degrade the pollutants in the wastewater. Hydrogen peroxide decomposes in one way or another, depending on the type of reaction and the system conditions [28]. Efficiency of H_2O_2 decreases with time due to the reasons that, it is consumed gradually and persistent intermediates remain in the system which hinders the performance of the system [26].

F. Integration of Treatment Processes

Integration of biological process with advanced oxidation processes was also evaluated in terms of COD and Color removal. Biotreated wastewater at optimized HRT (4 hrs) was ozonated for 6, 12, 18, 24 and 30 minutes followed by UV exposure for 10 to 60 min with three H_2O_2 doses (0.1, 0.5 and 1.0 mg/L) separately.

Fig. 5. Reduction in COD and color with different concentration of H_2O_2

1) Chemical Oxygen Demand

Maximum COD reduction with interaction of 6 min

ozonation and 60 min UV exposure in the presence of 0.5 ml/L H_2O_2 was up to 24.3 % followed by 20.6 and 15.0 % with 1.0 and 0.1 ml/L H_2O_2 dose respectively. Increase in the ozonation time significantly increased the COD reduction while keeping other conditions constant. Chemical oxygen demand decreased faster with 12 min of ozonation along with successive UV doses in the presence of three levels of hydrogen peroxide. The maximum COD reduction was 36.2 % however 21.7 % was achieved in just initial 10 minutes. Increase in ozonation time to 18 minutes, COD reduction jumped to 43, 75 and 89 % with 0.1, 0.5 and 1.0 ml/L H_2O_2 dose respectively after 60 minutes of UV irradiation time. Fastest COD reduction was observed with maximum dose of H_2O_2 (1.0 mg/L).

Most importantly, 54 % COD was reduced in first ten minutes of UV exposure in the presence of 1.0 mg/L H_2O_2 with the rate of 21 mg/min. However, this rate tremendously decreased to 3 mg/min after first ten minutes of UV exposure. Similar trend was observed in case of 0.5 mg/L H_2O_2 dose but with slightly less rate. Least COD was removed with 0.1 mg/L H_2O_2 dose at the rate of 3 mg/min.

Further increase in the ozonation time also increased the COD reduction. Maximum 93.4 % COD was reduced with 24 minutes of ozonation, 60 minutes of UV exposure and 1.0 mg/L H_2O_2 dose.

At 30 minutes of ozonation time followed by the 60 minutes of UV exposure in the presence of 0.5 mg/L H_2O_2 resulted 98 % COD removal (Fig. 6). All the treatment above 18 minutes of ozonation yielded COD within the permissible range i.e. 150 mg/L.

Increase in H_2O_2 dose increase the $\text{OH}\cdot$ radical production causes COD to reduce. Faster reduction in COD during first stage of oxidation was probably due to presence of sufficient amount of hydrogen peroxide which form $\text{OH}\cdot$ radicals and ultimately result in fair reduction of COD as per following equation.

COD reduction is also influenced by O_3 and UV irradiation exposure which act as a scavenger of organic material. Decrease in COD removal efficiency could be due to magnesium and calcium salts that crystallize on the quartz sheaths that enclose UV lamps submerged in the wastewater. The resulting scale can reduce the intensity of the light and shield the water from the UV light exposure [29]. This can also be due to increase in temperature that affects H_2O_2 solubility in wastewater and decreases the COD removal [30]. Hydrogen peroxide has also scavenging effect on $\text{OH}\cdot$ radicals and persistent compounds require more retention time for complete degradation.

2) Color

High efficiency was observed for color removal with integrated wastewater treatment system. Most of the treatments combination yielded above 90 % color reduction with maximum 99.7 %. Low doses of ozonation required longer UV exposure and high oxidation doses of H_2O_2 for

maximum color removal as shown in the Fig. 7.

Fig. 6. COD removal of 30 minutes pre ozonated and 4 hr bio treated WW with increase in UV time and H_2O_2 oxidation

Fig. 7. Color removal of 12 minutes pre ozonated and 4 hr biotreated WW with increase in UV time and H_2O_2 oxidation

Almost hundred percent colors were removed with the application of ozone above 12 minutes with 10 minutes of UV exposure and H_2O_2 doses.

Treatment of wastewater had two stages for color reduction. During the startup phase color removal was very high. After ten min color removal was slowed, % which could be due to the two possible reasons; first that wastewater had readily degradable compounds which require less time to degrade and, secondly, higher energy and time is required to degrade the remaining pollutants in wastewater because they are persistent.

High color removal than COD removal, was probably due to the partial oxidation of unsaturated bonds of chromophoric group responsible for color [31]. However, partially oxidized products contribute to COD because they still demand oxidation until their final degradation occurs [32].

This could also be attributed that H_2O_2 solubility decreases at elevated temperature which led to the decrease $OH^\bullet$ radicals and ultimately process efficiency decreased. The efficiency of the integration is higher due the presence of more number of oxidizing species in the system as compared to the individual process [26], [33] and heat produced by UV lamp as well as vigorous mixing of water [34].

IV. CONCLUSION

The study concluded that different wastewater treatment processes have varying degree of decoloration and COD reduction potential however, the efficiency of the process strongly depends on both the oxidation techniques used and the nature of the pollutants in the wastewater. Integration of AOPs with ASP yielded 98 and 100 % COD and Color removal.

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