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Performance of Combined Adsorption and Biological Process in Decolorization and Demineralization of Dye Wastewater

P. Anantha Narayanan, Ishwarya V., Mukesh Goel† and Ashutosh Das
Centre for Environmental Engineering, PRIST University, Thanjavur-613403, Tamil Nadu, India
†Corresponding author: Mukesh Goel

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ABSTRACT

This work studies the performance of combined adsorption and biological processes for degradation of crystal violet (CV) in wastewaters. The results were compared with biodegradation experiments conducted without adsorption. Activated rice husk was used as an adsorbent whereas, mixed culture was used for biological experiments. Effect of glucose concentrations and initial concentrations of dye were studied in detail. It was observed that combined degradation was very effective in removing CV from wastewaters. Adsorption with rice husk significantly increased the mineralization capacity of pollutants. Mineralization of actual textile effluents from textile industry also revealed the superior nature of combined treatment in comparison to biodegradation of dyes in wastewater.

INTRODUCTION

Industrial wastewaters tend to carry a huge load of organic and inorganic pollutants of which organic pollutants not only appear at high concentrations, but also exhibit a wide diversity with respect to their molecular structures (Wang et al. 2008, Venkata Mohan et al. 2013, Balapure et al. 2015). There are several techniques for the treatment of effluents, such as incineration, biological treatment, absorption onto solid matrices, etc. However, these techniques have their drawbacks, such as the formation of dioxins and furans, caused by incomplete combustion during incineration; long periods for biological treatment to have an effect, as also the adsorptive process, that is based on the phase transfer of contaminants without actually destroying them. In physical treatment methods, photocatalytic processes using TiO_2 are considered one of the most attractive methods for the treatment of wastewater because they offer a highly reactive, non-selective oxidant, that is hydroxyl radical (OH) destroying almost every pollutant present in the wastewater (Goel et al. 2010, Mohapatra et al. 2013, Nasar 2010, Subramaniam et al. 2016). The problem is further aggravated in the textile industry effluents, due to the complexity of their make-up. Thus, it can be seen that processes are being used that are not entirely appropriate for the treatment of textile effluents, thereby creating a major challenge for the industry and laundries that need to adapt to current regulations for the control of the colour of effluents with a high organic load.

Dyes are used in different industries like textile, cellulose, paper, handloom etc. After use, the dye wastes are discharged over soil or water bodies causing pollution. Due to toxicity of these dye wastes, it is necessary to treat them (Forgacs et al. 2004, Przystaœ et al. 2012, Dos Santos et al. 2007).

The use of filtration membranes and/or separation (Walsh 2001), in addition to incineration processes involving adsorption onto solid matrices, has also been adopted by the textile industry and is receiving considerable attention. However, all these processes only involve phase transfer, generating large amounts of sludge deposited at the end of the tanks and low efficiency in colour removal and reduction of the organic load. According to this scenario, many studies have been carried out with the aim of developing new technologies capable of minimizing the volume and toxicity of industrial effluents. Unfortunately, the applicability of these types of system is subject to the development of modified procedures and the establishment of effluent recycling systems, activities that imply evolutionary technologies and which are not yet universally available. Thus, the study of new alternatives for the treatment of many industrial effluents currently produced is still one of the main weapons to combat the phenomenon of anthropogenic contamination (Han et al. 2016, Dharajiya et al. 2016).

The aim of the present work is to explore the potential of activated rice husk being utilized as adsorbent in the combined biological and adsorptive removal of crystal violet (CV) from its aqueous solution. Mixed culture obtained from the sewage treatment plant was used for biological treatment.

MATERIALS AND METHODS

The water-soluble CV was obtained from M/s Merck and its stock solution was prepared in double distilled water. Solutions of desired concentrations of the adsorbate were prepared from stock solution, and double distilled water was used for necessary dilutions. All reagents used in the investigation were of analytical grade. Fresh stock solution, as required, was prepared every day and stored in a brown colour glass reservoir of 5 L capacity to prevent photo-oxidation. The initial concentration of dye (C_0) was ascertained before the start of each experimental run by using UV spectrophotometer.

Culture collection: Mixed culture was obtained from a nearby sewage treatment plant. The cultures were maintained at 4°C in nutrient broth. The cultures were transformed into continuous subcultures for every 10 days. The medium composition used throughout the study for biological treatment is: yeast extract (0.34 g/L), NH_4NH_3 (0.84 g/L), KH_2PO_4 (0.134 g/L), K_2HPO_4 (0.234 g/L), $MgCl_2.6H_2O$ and varying concentrations of glucose.

Preparation of activated rice husk: Rice husk was collected and dried for overnight at 105°C in an electric oven. The dried rice husk was activated with 1:2 H_3PO_4. The activation method involved the use of 100 g of rice husk with 120 mL H_3PO_4 mixed and kept overnight at room temperature. It was then activated at 500°C for 2 hours in nitrogen atmosphere. The samples so generated were washed with distilled water (until getting pH 7), dried and sieved (at 125 microns) for use in adsorption studies.

Measurement of chemical oxygen demand: Chemical oxygen demand was measured using HACH colorimeter (DR 890). The COD solution (HR grade 0-1500 ppm) was prepared by mixing 0.25 mL of COD solution A and 2.8 mL of COD solution B. To this solution, 2 mL of centrifuged sample (include dilution) was added. The digestion was done at 150°C in HACH COD digester for two hours using HACH COD vials. Final COD value after air cooling is taken in HACH-DR/890 colorimeter (Program No 17).

Experimental Procedure

Adsorption: For each study, a 100 mL synthetic dye solution of 400 ppm concentration was prepared in 250 mL Erlenmeyer flask and a known amount of adsorbent was added to each bottle at pH 4. The solution was equilibrated for 4 h at room temperature, followed by filtration of the adsorbent and subsequent analysis of filtrate for dye concentrations. The percentage removal of dye was calculated using the following relationship:

$$\% \text{ Removal} = \frac{100(C_0 - C_e)}{C_0},$$

Where, Ce is the equilibrium adsorbate concentration (ppm) and C_0 is initial concentration (ppm).

Integrated experiments: The adsorbed wastewater was subjected to combined treatment using biological degradation. An appropriate quantity of inoculums and media were added to adsorbed wastewater to make up the reactor volume to 30 mL. Biological treatment for dye removal was conducted using 100 mL conical flask. The reactor was maintained at desired temperature in orbital shaker. pH 7 was maintained throughout the experiments by adding NaOH or H_2SO_4. Both the COD and dye concentrations were measured every 12 hours. The experiments were conducted for six different initial concentrations of dye (100, 200, 300, 400, 500 and 600 ppm). Three different glucose concentrations (1 g/L, 2 g/L and 3 g/L) and four different temperatures (25, 30, 35 and 40°C) were used for the biological experiments. The experiments were also conducted for cultures without glucose. The experiments were run for 7 days. All measurements were done in duplicate. The inoculums' concentration was fixed at 15%.

RESULTS AND DISCUSSION

Present experiments were conducted to find out the optimum adsorbent concentration for dye removal. Dye with 400 ppm inlet concentration was subjected to adsorption using activated rice husk. The results are presented in Fig. 1. It can be seen that high adsorbent dosage enhance the dye removal. During four hours of adsorption, 39% removal was observed for 1 g/L dosage which increased to 58% for 5 g/L TiO_2. However, removal did not show further enhancement when the adsorbent dosage was increased beyond 5 g/L. Though increased dosage of adsorbent provide more surface area for adsorption to take place, above a certain value of the adsorbent, adsorption is independent of adsorbent dosage. Thus, 5 g/L of activated rice husk was used for all the integrated treatment experiments.

Integrated treatment: With an objective to achieve further removal, adsorbed dye wastewater was subjected to biological degradation. Both the dye removal and COD removal were noted for the experiments. We studied the effect of glucose concentration and initial concentration for this work. Besides, the effect of temperature was also studied.

Effect of glucose: Figs. 2 and 3 show the dye removal for both, integrated treatment and biodegradation alone at different glucose concentrations (0, 1 and 2 g/L) for 400 ppm

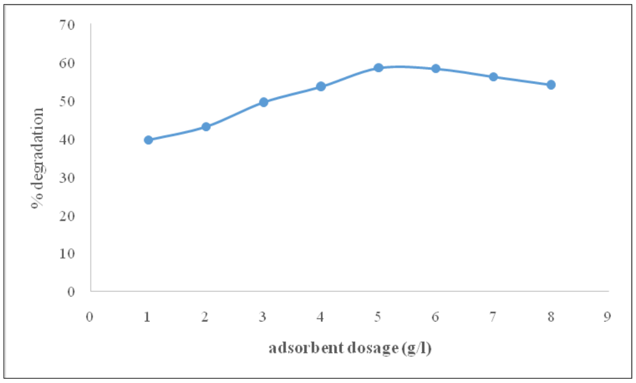

Fig. 1: Effect of adsorbent dosage on adsorptive removal of CV from wastewater.

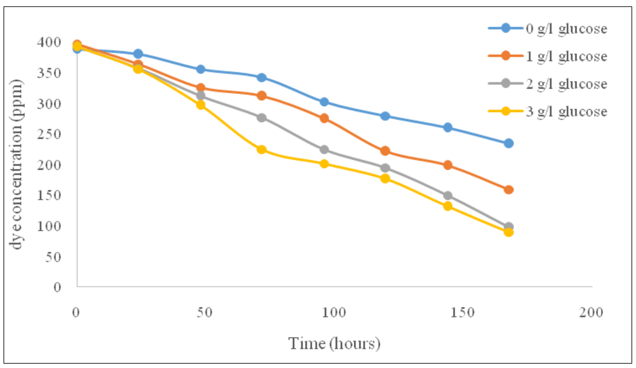

Fig. 2: Biodegradation of CV at different glucose concentrations without adsorption.

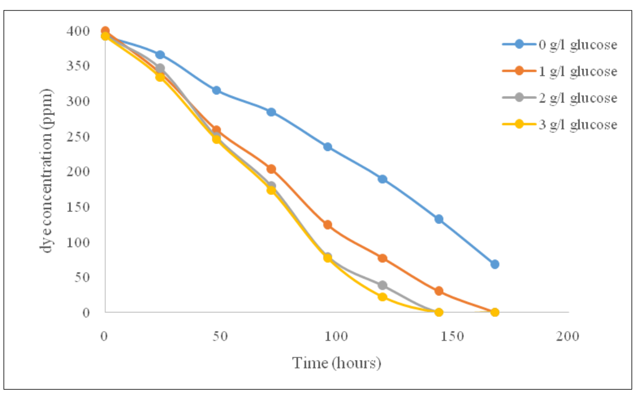

Fig. 3: CV reduction for combined treatment at different glucose concentrations with 4 hours of adsorption.

of dye. The results were observed for one week. For biodegradation alone, effect of glucose was more prominent than that of combined treatment. Increase in glucose concentration steadily increased the degradation rate of dye. The dye degradation increased from 47% to 89% as glucose concentration increased from 1 g/L to 3 g/L (Fig. 2). In case of pretreatment with adsorption, complete dye removal was observed in all the cultures, however, the cultures with 2 g/L and 3 g/L glucose degraded the dye within 96 hours (four days), whereas cultures without glucose yielded the complete removal only by the end of the week for all the concentrations. It was less affected by increase in glucose concentration from 2 g/L to 3 g/L. The kinetic data show that both the glucose concentrations (2 g/L and 3 g/L) yielded the similar trends in degrading dye (Fig. 3).

The results were further confirmed by COD removal data. COD removal was strongly influenced by the presence of glucose for biodegradation alone, as shown in Fig. 4. On the contrary, combined treatment showed much more rigidity with respect to the presence of glucose as observed for dye degradation. Adsorption with rice husk significantly increased the mineralization capacity of the pollutants. This is an important finding as it has the potential to replace expensive biogenic substrate with low cost rice husk, thus making the process much more economical.

Effect of initial concentration of dye: Biological degradations were carried out at different initial concentrations with and without adsorbed solution. Biological degradation without adsorption is presented in Fig. 5. The figure shows that for the lowest concentration (100 ppm), complete degradation took six days, whereas degradation for highest concentration (600 ppm) was incomplete by the end of the week. Only 73% removal was observed. On the other hand, the process combination was much more efficient and effective in biological degradation of dyes (Fig. 6). It can be seen that for an initial concentration of 100 ppm dye, complete degradation was achieved in four days, 33% less compared to biodegradation alone. And culture with 600 ppm initial concentration got completely degraded in seven days. It can be thus noted that integrated treatment is very effective as the concentration of pollutant increases. For example, culture with 300 ppm initial concentration resulted in 92% removal by five days in integrated treatment, whereas the degradation rate was only 57% with biodegradation alone. Thus, integrated treatment is 43% more efficient compared to biodegradation alone.

COD removal rate data show the same trend. COD removal rate is detailed in Table 1 for both, biodegradation alone and integrated treatment. Integrated treatment yielded in complete COD removal for cultures with 100 ppm concentration. However, biodegradation alone could result in only 66% removal. Similarly COD removal for higher concentrations (600 ppm) is efficiently degraded by integrated treatment than biodegradation alone.

Effect of temperature: Since temperature is an important parameter in chemical/biochemical reactions, we studied the effect of temperature on both, biodegradation as well as integrated treatment of dye. It can be seen from Fig. 7 that

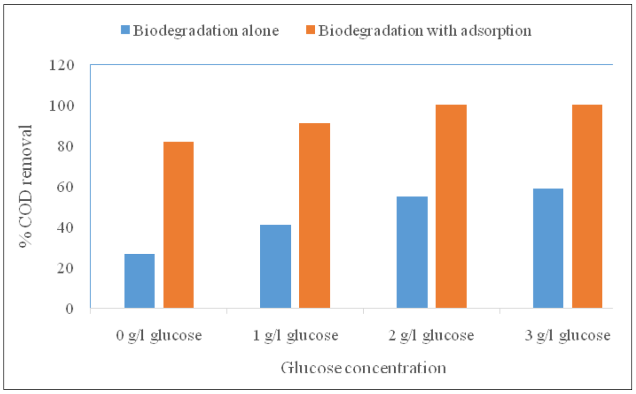

Fig. 4: Demineralization data for combined treatment of CV after 4 hours of adsorption and without adsorption for all glucose concentrations.

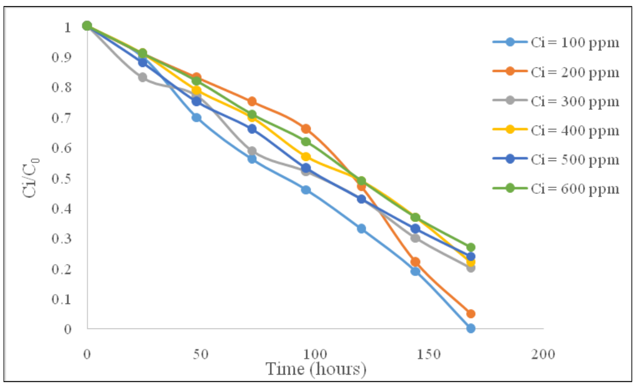

Fig. 5: Biodegradation of CV at different initial concentrations without adsorption.

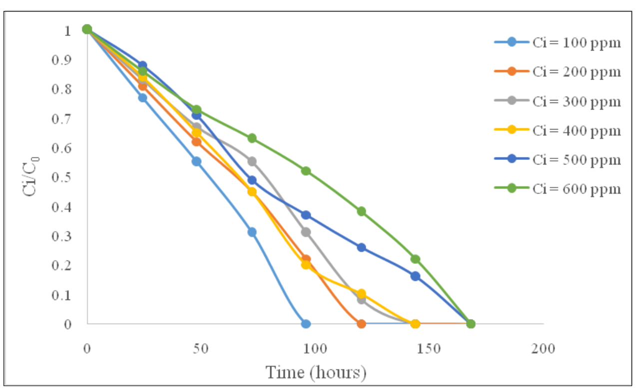

Fig. 6: CV reduction for combined treatment at different initial concentrations with 4 hours of adsorption.

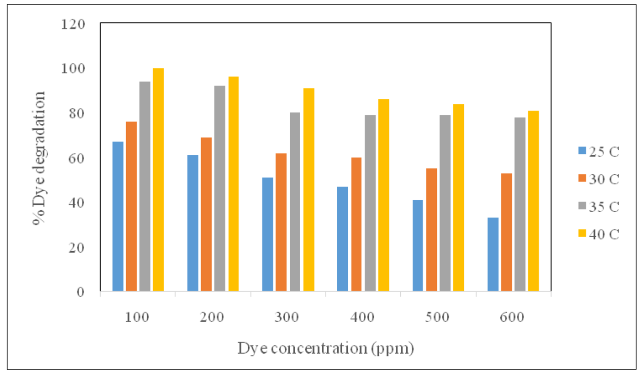

Fig. 7: Effect of temperature on biodegradation of CV without adsorption.

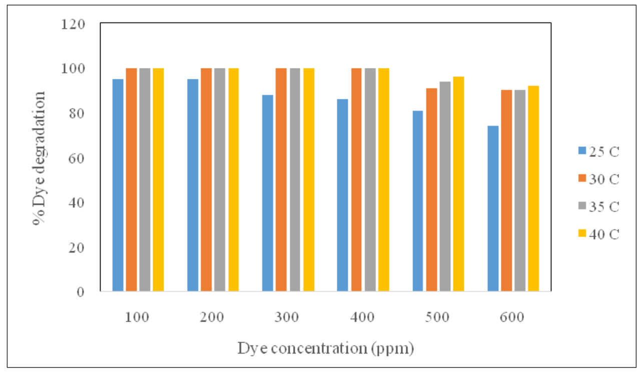

Fig. 8: Effect of temperature on biodegradation of CV with 4 hours of adsorption.

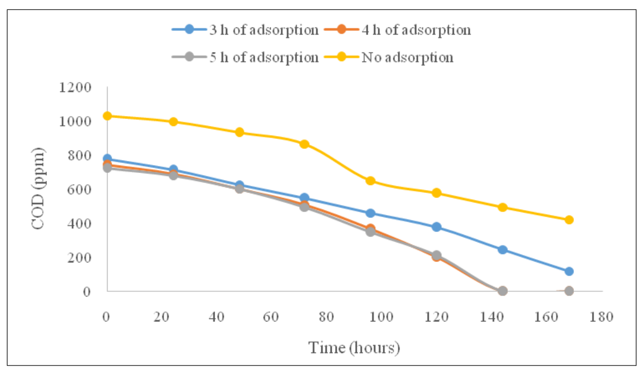

Fig. 9: Demineralization of textile effluents for combined treatment of CV after 3 h, 4 h and 5 h without adsorption.

dye degradation increased with the increase in temperature. At 25°C, only 67% removal was observed which increased to 100% at 40°C for biodegradation alone for 100 ppm of dye. Similarly, integrated treatment results showed increased degradation with the increase in temperature (Fig. 8). High temperature, however, necessitates high operating cost and hence actual plant design normally involves trade off in order to accomplish the best wastewater treatment options.

Mineralization of textile effluents: Actual textile effluent was obtained from nearby industry with an initial COD of 1065 ppm and subjected to mineralization study for both, biodegradation as well as integrated treatment. The experiments were carried out with 4 g/L of activated rice husk at 30°C. We also wanted to verify the effective time of adsorption and hence integrated treatment was conducted after 3

Table 1: Effect of the dye concentration on COD removal of dye wastewater.

Dye concentration (ppm)	Biodegradation alone glucose concentration				Biodegradation with adsorption glucose concentration			
	0 g/L	1 g/L	2 g/L	3 g/L	0 g/L	1 g/L	2 g/L	3 g/L
100	55	57	65	69	93	100	100	100
200	52	55	65	66	89	100	100	100
300	41	49	56	66	86	93	100	100
400	27	41	55	59	82	91	100	100
500	25	33	51	53	77	87	91	94
600	21	30	43	46	72	85	90	90

hours and 5 hours of adsorption. Results are illustrated in Fig. 9. It can be seen that mineralization followed the similar trend as synthetic dye wastewater. The results evidently indicate that adsorption had improved biodegradability of effluents. Complete mineralization was accomplished in six days after four hours of adsorption. Increase in the adsorption time to 5 hours did not significantly improve the mineralization rate, whereas biodegradation after 3 hours of adsorption resulted in only 89% of COD removal. In contrast to integrated treatment, biodegradation alone yielded poor performance and only 59% of mineralization was observed. Therefore, a two-step process consisting of adsorption and biological degradation is more effective for treating textile effluents. Pretreatment with a suitable process considerably reduced the time required for complete mineralization.

CONCLUSION

The present study effectively degrades the dye wastewater containing crystal violet (CV) using combined adsorption and biological process. Effects of glucose concentration and initial concentration of dyes were studied in detail. It was found that combined degradation did not show much importance to glucose concentration as it increased from 2 g/L to 3 g/L, whereas biodegradation was affected by increase in glucose concentration. The dye degradation increased from 47% to 89% as glucose concentration increased from 1 g/L to 3 g/L in case of biodegradation. Furthermore, integrated treatment is very effective as the concentration of pollutant increases. Experiments with actual textile effluents also showed that adsorption had improved biodegradability of effluents; complete mineralization was accomplished in six days. On the other hand, biodegradation alone yielded poor performance and only 59% of mineralization was observed.

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