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Research Article

EFFICIENCY OF BIOLOGICAL AND ADVANCED OXIDATION PROCESS IN REMEDIATION OF TEXTILE INDUSTRY DYE BATH

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ABSTRACT

Textile dye bath released by dyeing industries contain higher pH, colour intensity and physicochemical parameters. The effluent for remediation studies was collected from Common Effluent Treatment Plant (CETP), Perundurai, Tamil Nadu, India. The rate of decolourization achieved in remediation of dye bath by bacterial treatment using *Lysinibacillus spheaericus* SK13 was 68%, chemical treatment using advanced oxidation processes was 63%, and their combined treatment processes was 75%. The bacterial remediation proved better in the reduction of effluent physicochemical parameters compared to chemical treatment processes. The remediation of dye bath using combined bacterial and chemical treatment decreased its pH to neutral, along with significant reduction of total suspended solids, ammonium, nitrate, nitrite, phosphate, chromium, copper, cobalt, zinc by 81-96%; turbidity, BOD, COD lead, manganese, cadmium, sodium and phenolic compounds by 70-79%; electrical conductivity, iron, phosphate, oil and grease by 60-68%; turbidity, total solids, total dissolved solids, bicarbonate, chloride by 40-49%; sulphate, potassium by 35-38%; and fluoride, silicate and magnesium by 12-20%. The evaporation of this treated dye bath in the evaporation tanks of CETP can highly reduce the secondary sludge production compared to that of untreated dye bath.

Keywords: Remediation, Advanced oxidation processes, *Lysinibacillus spheaericus*, Common Effluent Treatment Plant.

INTRODUCTION

Textile dye bath are highly coloured containing highly reactive azo dyes and complex structured polymers that had not reacted with the fiber during textile processing (Muhammad *et al.*, 2008). Along with hydrolyzed reactive and disperse dyes, higher concentration of sodium chloride and soda ash are present (Balamurugan *et al.*, 2011). Along with these carcinogenic dye ingredients, dye bath contains higher levels of physical parameters and metal ions. This high strength textile dye bath is highly challenging to degrade using most treatment processes (Manekar *et al.*, 2014). In the Common Effluent Treatment Plant (CETP), Perundurai, Erode District, Tamil Nadu India, the dye bath treatment is usually evaporated from evaporation tanks, using heat exchangers. The recovered water is further reused for textile processing. The sludge deposited is stored forever without any possibility for proper disposal and reuse (Ramesh *et al.,* 2009). The coloured effluent will pollute the aquatic environment, and in addition the added colour will affect light penetration in river waters affecting photosynthesis and imbalance in the ecosystem (Balamurugan and Kannadasan, 2012). The bacteria can decolourize dye bath with production of least sludge in more effective and economical way compared to other treatment processes. *Lysinibacillus sphaericus* used in this study is a competent bacterium, capable of effective degradation of textile dyes (Senthil Kumar *et al.,* 2016), by the breakage of azo linkages in the textile dyes present in the dye bath. The bacteria can effectively degrade the dyes using the enzyme azoreductase, under anaerobic conditions at normal temperature, hence chosen for remediation of remediation of dye bath (Forgacs *et al.,* 2004). The aromatic simpler amines formed after bacterial remediation processes and highly reactive electrophilic diazonium salts are difficult to be removed and degraded further from the dye bath (Saratale *et al.*, 2011). They are toxic on release into natural environment and its accumulation into any aquatic organisms will be highly mutagenic affecting food chain (Mahbub *et al.*, 2012).Chemical remediation using Advanced Oxidation Process (AOP; with H_2O_2), can increase the rate of decolourization of dye bath. The highly reactive radicals [HO•] produced through photocatalytic decomposition of H_2O_2, can react with the azo dyes and other organic components present in the dye bath into CO_2 and H_2O (Muhammad *et al.,* 2008). The chemical treatment can be combined after bacterial treatment process to increase the rate in decolourization and effectively decrease the physicochemical parameters of the dye bath with reduced secondary sludge.

MATERIALS AND METHODS

Necessary volume of textile dye bath was collected from CETP, Perundurai. The pH was determined at the sampling site, before transported to laboratory. The bacterial, chemical and its combined treatment processes were optimized and pursued in the laboratory in 10 L volume tanks.

Treatment Methodologies

For bacterial treatment of the dye bath, *Lysinibacillus sphaericus SK* 13 (NCBI Accession No. KF032717) was used for the purpose of remediation of textile dye bath. This bacterium was isolated from the soil contaminated with textile dye effluent at the disposal site. It was screened for its ability to degrade textile dye effluent, and the bacterium was maintained in Luria Bertani broth under anaerobic condition at 25ºC and pH 8. About 50 mL of 24 hour culture with an optical density of 1.0 at 600 nm was added to one litre of dye bath and maintained in anoxic condition for 48h hours (Senthil *et al.,* 2016). The tanks were covered tightly with plastic sheet for maintenance of anoxic conditions. The decolourized effluent was centrifuged at 10,000 rpm for 10 min to remove the cell mass from it. The chemical treatment of textile dye bath using AOP (Oller *et al.*, 2011), was optimized with 0.49M of H_2O_2 (50 mL of H_2O_2; 30% w/w) was added to one litre of RE, and incubated in direct sunlight for 16 ± 2 hours at 1,14,000 ± 15,000 illuminance (Lux). The treated effluent was centrifuged at 5,000 rpm for 10 min to remove the sludge. The treatment tank was tightly covered with transparent (for photocatalytic reaction) plastic sheets to limit the rate of evaporation. The bacterial treatment processes were further combined with chemical treatment processes for increased improvement of effluent physicochemical parameters.

Decolourization level

UV-Vis Spectrophotometric analysis (Shimadzu UV-1800) was performed at regular intervals of 24 hours to measure the decolourization percentage of the treated effluent, following the method adopted by ADMI (American Dye Manufacturers Institute), and recorded in ADMI values (Khandare and Govindwar, 2015). The colour removal was calculated as:

ADMI removal % = (Initial ADMI - Final ADMI/ Initial ADMI) x 100

Analysis of effluent parameters

The pH of the samples was determined using standard pH meter. The physicochemical parameters of turbidity, Total Dissolved Solids (TDS), Total Suspended Solids (TSS), Electrical Conductivity (EC) and Total Solids (TS) were determined following the Standard Method for the Examination of Water and Wastewater (American Public Health Association, 2005). The Biological Oxygen Demand (BOD) was determined by titration, method, while that of Chemical Oxygen Demand (COD) by Open Reflux method. Sodium and potassium ions were determined by Flame Photometer method. Phosphate, nitrate, nitrate, silicate, chloride (Mohr's), ammonia (sulphuric acid titration), bicarbonate and carbonate were determined by titration method, sulphate by Gravimetric method, concentrations of heavy metals (calcium, magnesium, iron, fluoride, manganese, copper, zinc, lead, cadmium, chromium, cobalt, mercury) using Atomic Absorption Spectroscopy (SIMADZU, Model-AA3800/G) (Hussein, 2013). Oil, grease and phenolic compounds were determined by chromatography following Rauckyte *et al.,* (2010). Significance of difference between the RE (control) and the treated groups of RE (results of experiments pursued in triplicates) were analyzed using One way ANOVA followed by Tukey's post hoc test, with p≤ level at 0.05 to check the interactive effects between different parameters.

RESULTS

The dye bath contains extremely high levels of pH, physicochemical parameters along with higher levels of metal ions especially chloride, sulphate, calcium, magnesium, sodium, oil, grease and phenolic compounds. It is highly toxic on release into natural environment. Dye bath after treatment by bacterial, chemical and its combined treatment processes showed 68, 64 and 75% rate of decolourization. All the treatment processes are capable of bringing the dye bath pH to neutral. The treatment of textile dye bath using bacterial, chemical and its combined treatment process include the significant decline in physical parameters of turbidity by 55, 46 and 70%; total solids by 38, 32 and 49%; total dissolved solids by 36, 29 and 47%; total suspended solids by 82, 78 and 92%; electrical conductivity by 61, 57 and 68%; BOD by 58, 54 and 79%; and COD by 58, 34 and 74%respectively. The decline in metal ions includes the decline in bicarbonate by 22, 12 and 44%; chloride by 27, 21 and 40%; sulphate by 18, 14 and 35%; fluoride by 12, 7 and 13%; silicate by 9, 5 and 12%; ammonium by 76, 63 and 81%; nitrate by 45, 23 and 84%; nitrite by 73, 34 and 86%; phosphate by 92, 92 and 96%; calcium by 68, 66 and 70%; sodium by 67, 66 and 73%; magnesium by 12, 8 and 20% and potassium by 31, 12 and 38%. The reduction in heavy metals includes the reduction of iron by 42, 27 and 60%; lead by 49, 17 and 70%; chromium by 63 , 30 and 80%; zinc by 79, 69 and 90%; manganese by 46, 39 and 71%; cadmium by 39, 9 and 70%; copper by 75, 50 and 87%; cobalt by 77, 22 and 89%; mercury by 66, 33 and 100%; further the decline of oil and grease by 66, 16 and 67%; and phenolic compounds by 48, 52 and 70%. The highest decline of effluent physical parameters and metal ions were achieved using combined bacterial and chemical treatment processes (Table 1).

Table 1. Decrease in the physico-chemical parameter of dye bath using chemical, bacterial and its combined treatment processes.

	Untreated Dye Bath	**Bacterial Remediation**	**Chemical Remediation**	**Bacterial + Chemical Remediation**
Decolourization(in %)	Nil	68 %	62%	75%
Physical parameters				
pH	11.16 ± 0.07	7.62 ± 0.06[a]	7.89 ± 0.04[a]	7.53 ± 0.02[a]
Turbidity	1750 ± 22.68	780± 8.19[a]	950 ± 11.67[a]	520 ± 7.26[a]
TS	19740 ± 38.22	12160 ± 7.11[a]	13480 ± 78.23[a]	10120 ± 2.54[a]
TDS	18870 ± 96.62	12100 ± 7.72[a]	13310 ± 2.05[a]	9980 ± 9.68[a]
TSS	770 ± 20.82	140 ± 5.81[a]	170 ± 4.41[a]	60 ± 9.45[a]
EC	4.87 ± 0.10	1.89 ± 0.11[a]	2.08 ± 0.50[a]	1.56 ± 0.11[a]
BOD	685 ± 26.03	285 ± 10.17[a]	316 ± 7.05[a]	143 ± 9.00[a]
COD	1236 ± 28.87	520 ± 10.14[a]	816 ± 13.30[a]	316 ± 8.08[a]
Metal anions and cations				
Bicarbonate	546 ± 8.88	426 ± 13.48[a]	482 ± 6.96[a]	308 ± 1.89[a]
Chloride	486 ± 4.98	353 ± 08.76[a]	386 ± 5.81[a]	289 ± 7.77[a]
Sulphate	136 ± 4.05	112 ± 10.09[a]	116 ± 6.08[a]	89 ± 6.24[a]
Fluoride	5.89 ± 0.08	5.16 ± 0.05[a]	5.49 ± 0.06	5.09 ± 0.07[a]
Silicate	3.88 ± 0.23	3.52 ± 0.13	3.69 ± 0.23	3.42 ± 0.15[a]
Ammonium	22 ± 0.41	5.20± 0.21[a]	8.23± 0.15[a]	4.14± 0.14[a]
Nitrate	15 ± 0.20	8.28± 0.11[a]	11.56± 0.11	2.36± 0.18[a]
Nitrite	8 ± 0.11	2.12± 0.02[a]	5.24± 0.18[a]	1.12 ± 0.17[a]
Phosphate	15 ± 0.12	1.10 ± 0.03[a]	1.21 ± 0.23	0.52 ± 0.14[a]

Calcium	1540 ± 46.38	486 ± 08.82[a]	520 ± 9.24[a]	456 ± 4.91[a]
Sodium	1689 ± 49.09	548 ± 11.59[a]	579 ± 14.5[a]	458 ± 06.2[a]
Magnesium	235 ± 05.49	206 ± 02.31[a]	215 ± 2.89	189 ± 10.33[a]
Potassium	0.32 ± 0.02	0.22 ± 0.01[a]	0.28 ± 0.01	0.20 ± 0.01[a]
Heavy metals				
Iron	0.89 ± 0.04	0.52 ± 0.02[a]	0.65 ± 0.04[a]	0.36 ± 0.03[a]
Lead	0.63 ± 0.06	0.32 ± 0.05[a]	0.52 ± 0.05	0.19 ± 0.02[a]
Chromium	0.59 ± 0.05	0.22 ± 0.03[a]	0.41 ± 0.03[a]	0.12 ± 0.02[a]
Zinc	0.39 ± 0.02	0.08 ± 0[a]	0.12 ± 0.01[a]	0.04 ± 0[a]
Manganese	0.28 ± 0.02	0.15 ± 0.01[a]	0.17 ± 0.01[a]	0.08 ± 0.01[a]
Cadmium	0.23 ± 0.05	0.14 ± 0.01[a]	0.21 ± 0.01	0.07 ± 0.01[a]
Copper	0.16 ± 0.01	0.04 ± 0[a]	0.08 ± 0.01[a]	0.02 ± 0[a]
Cobalt	0.09 ± 0.01	0.02 ± 0[a]	0.07 ± 0.01[a]	0.01 ± 0[a]
Mercury	0.003 ± 0	0.001 ± 0[a]	0.002 ± 0[a]	0[a]
Other parameters				
Oil and Grease	0.06 ± 0.02	0.02 ± 0[a]	0.05 ± 0	0.02 ± 0[a]
Phenolic ompounds	1.08 ± 0.01	0.52 ± 0.01[a]	0.56 ± 0.01[a]	0.32 ± 0.01[a]

Values are expressed as mean±SE. Turbidity units expressed in NTU, Electrical conductivity is measured in dsm^{-1}, and units of other parameters except pH are expressed in mg/L. The letter [a]represents significant difference among treatment groups when compared against untreated effluent at $p < 0.05$.

DISCUSSION

The bacteria had decolourized the effluent by breaking the azo linkages in the textile dyes present in the dye bath under anaerobic conditions (Nachiyar *et al.*, 2014). But still the secondary aromatic intermediatory compounds formed after the bacterial remediation of dye bath are not completely degraded in bacterial remediation (Ogugbue and Sawidis, 2011). Chemical remediation using AOP can degrade the aromatic and chlorinated hydrocarbons present in the dye bath (Oller*et al.*, 2011), as well as the intermediatory products produced after bacterial remediation into secondary non-degradable low molecular weight intermediates (Rodrigues *et al.*, 2014). The chemical treatment is combined after bacterial treatment, in order to treat the xenobiotic and other toxic non-degradable compounds cannot be degraded by bacterial remediation (Mahvi *et al.*, 2012). Hence the combined bacterial and chemical treatment processes can be useful for the increased removal of physiochemical parameters and metal ions from the dye bath, in order to decrease the production of sludge produced after the heating process in the CETP.

CONCLUSION

The treatment of dye bath using combined bacterial and chemical treatment processes can efficiently decrease its physical parameters and metal ions, than their single use in remediation processes. The reduced physicochemical parameters in the dye bath will produce less sludge after it is evaporated in the evaporation tanks using heat exchangers, compared with the untreated dye bath. Thus the reduced secondary sludge produced will reduce the space constraints required for its storage.

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