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Decolorization of Tartrazine azo dye by Sulphidogenesis Process and the effect of pH

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Abstract—This study aims to investigate Tartrazine as a model azo dye with a concentration of 100 mg/l for decolorisation by sulphidogenesis process in a sequencing batch reactor (SBR). SBR was operated at 30±2 ℃ room temperature and initial cycle time of 7 days which has later been reduced to 1-2 days. SBR was operated at COD/Sulphate ratio of 1. SBR did well in terms of reducing color, COD and sulphates. Maximum color, COD and sulphate removal were 84%, 74% and 88% respectively. Activated sludge could be used as seed biomass for the development of the process. In the later part of our study, we tried to optimize the best pH to remove color by setting the six reactors with varying pH. The results show that optimal pH for decolorization is between 8 and 9 and effective color removal (87-92%) could be achieved in a day. It is also seen that alkaline pH is more favoured than acidic pH for decolorization of Tartrazine. The results of this study indicate an economical and ecofriendly technique for decolorization of Tartrazine at 100 mg/l.

Key words: Sulphidogenesis, Sulphate reducing bacteria (SRB), Sequencing batch reactor (SBR), Acclimatized sludge, Anoxic

I. INTRODUCTION

Azo dyes are aromatic compounds that consist of –N=N- groups. They are the major class of synthetic dyes used in various industries, namely, textile, food coloring, cosmetics and paper printing, and among these textile industries largest user of dyes. All the dye does not stick to the fabric and depends on the dye used, the loss could vary from 2% for basic dyes to as high as 50% [1] for reactive dyes and upon their release into the environment, they pose a severe threat. These dyes and their by-products are carcinogenic in nature and are reported as more threatening wastewaters when compared to other industrial effluents, even their low concentrations equal to "1 mg/l" can make surface water looks colorful, which is not good aesthetically and also that color interrupts the sunlight which inhibit photosynthesis process thus affects the aquatic life [2]. Color removal from wastewaters has been a big challenge over the last years. There are many physical and chemical processes to treat the colored effluents, but their operation cost is quite high, while the biological process offers a lower cost and an eco-friendly technique [3].

Generally, mineralization of azo dye requires both anaerobic and aerobic biological processes [4]. Though the anaerobic process removes color and COD and formation of aromatic amines, these are not mineralized in anaerobic phase and are considered toxic in nature [5]. This tends to develop a 2 stage sequencing anaerobic-aerobic batch process; reduced products are oxidized in the second phase. The role of sulphidogenesis process may be effective for decolorisation of azodyes since sulphate concentration can be in the range 1700-2700 mg/l in the textile dyeing effluents [6]. Ozdemir, et. al. [7] investigated the performance of azo dye reduction in a sulphidogenic anaerobic reactor over a period of 400 days. The reactor consists of 4 sections, in which three of them are anoxic and the fourth section is aerated. The dye used in this study was C. I Reactive Violet 5 and azo dye concentrations varied between 20 and 200 mg/l, Throughout the study the COD/SO_4^{2-} ratio was less than 1 ensuring sulphidogenic conditions. The results in this study showed high COD removal efficiencies and high dye removal efficiencies. Also, as the COD/SO_4^{2-} ratio was increased, there was increased sulfate removal efficiencies from 81% to 91 %. The maximum COD removal, Sulfate removal and color removal efficiencies were 98%, 98% and 93% respectively.

So far, different dyes with different anaerobic reactors for textile wastewaters have been used; namely, fluidized bed reactors [8], upflow sludge

blanket reactor and packed bed reactors [9], anaerobic baffled reactor [7]. However, Tartrazine has not been examined under sequencing batch reactor (SBR) with sulfate reducing bacteria (SRB), so this study focuses on decolorization of Tartrazine dye sulphate reducing bacteria in SBR process and the effect of pH on decolorisation was investigated.

II. MATERIALS AND METHODS

A. Nutrient media

The following nutrients in g/l were used for SBR feed: Yeast, 1; $NaHCO_3$, 2.1; NH_4Cl, 0.11965; K_2HPO_4, 0.035; $MgCl_2$, 0.2; H_3BO3, 0.0005; $ZnCl_2$, 0.0005; $(NH_4)_6Mo_7O_{24}.4H_2o$, 0.0005; $NiCl_2.6H2O$, 0.0005; $AlCl_3.6H_2O$, 0.0005; $MnCl_2.4H_2O$, 0.0005; $CoCl_2.4H_2O$, 0.0005; $Na_2SeO_3.5H_2o$, 0.001; $CuSO_4.5H_2O$, 0.0005; $CaCl_2$, 0.003; $FeCl_2$, 0.000015; EDTA, 0.000015.

B. Electron sources

Sucrose (2.5 g/l) is used as readily degradable carbon source, at the same time it provides electron for the cleavage of azo dye. Tartrazine dye in the concentration of 100 mg/l is used for colored wastewater in each cycle of SBR operation. The electron acceptor in the experiment was sulphate (added as Na_2SO_4, 3.7 g/l).

C. Reactor setup and experimental design

To start the SBR to check the feasibility of decolorisation of Tartrazine, we took the available acclimatized sludge in the Environmental Engineering laboratory which was used for decolorisation of Methyl orange by sulphidogenesis process. Batch incubations were conducted at 30±2°C in glass serum bottles of "150 ml" capacity sealed with neoprene rubber septum and secured with aluminium caps. We took three serum bottles; 1 control with just dye and nutrient media while 2 bottles of nutrient media, dye of 100 mg/l concentration and sludge (25 ml) in each bottle. The COD/SO_4^{2-} ratio of the nutrient media was kept as one to promote the sulphidogensis by SRB [10]. These sealed bottles were then kept on an orbital shaker at 120 ± 2 rpm (Scigenics Biotech, India) with initial cycle time of 7 days. The cycle time reduced in subsequent cycles as and when the decolorization happened. The time for filling, settling and decanting was ≤ 1 h in each cycle.

After seeing the feasibility of decolorization, an enrichment of activated sludge which was collected from VIT Chennai wastewater treatment plant with MLVSS value of 1.344 g/L was done in a 5 l SBR (Plastic cargo) to decolorize 100 mg/l of Tartrazine with 1000-1200 ml of sludge and rest of the reactor filled with nutrient media at COD/SO_4^{2-} ratio equal to one. The nutrient media was replaced every 6 days interval and on acclimatization the cycle time reduced to 2 days. The time for filling, settling and decanting was ≤ 2 h in each cycle. After 10 cycles of acclimatization the sludge was drawn to study the effect of pH in decolorization. For this 6 serum bottles with 30 ml of acclimatized sludge (MLVSS = 3.479 g/l) in each bottle was taken and then each bottle was filled with Tartrazine dye of concentration 100 mg/l in nutrient media. The nutrient media with dye was pre-adjusted with varying pH of 5, 6, 7, 8 and 9 with 1N HCl and 1N NaOH and sixth bottle was used as a control with just acclimatized sludge and dye concentration of 100 mg/l and pH 7.6±0.1. These sealed bottles were then kept on an orbital shaker at 120 ± 2 rpm.

III. ANALYTICAL PROCEDURES

Color absorbance, sulphate, sulfide, soluble COD and pH were measured after each cycle. Oxidation Reduction Potential (ORP) was also measured weekly to ensure the anoxic conditions in the reactors. Samples were centrifuged for 10 minutes before analyzing absorbance, COD and sulphates while pH and sulfide were measured with MLSS samples without centrifugation.

Color was measured using UV-VIS Spectrophotometer, and absorbance was measured at the peak absorption wavelength in visible range (λ_{max} = 425nm) against a baseline defined by triple distilled water. All physicochemical parameters were measured according to the standard procedures [11]. ORP was measured with an ORP electrode (platinum with silver/silver chloride as electrode, 3M KCl) connected to digital meter. pH was measured using pH of 2-point pH meter. Sulphate was measured as absorbance at "420 nm" wavelength followed by experimental derived curve (R^2=0.994). Sulfide analysis is done at the end of each cycle by the iodometric method mentioned in APHA 1998 [11]. All the tests and values were taken in duplicates and average values are reported.

IV. RESULTS AND DISCUSSION

A. Color removal

The sequencing batch reactors were initially operated with a cycle time of 7 days and after each cycle, fresh nutrient media with dye concentration of 100 mg/l was added for subsequent process to induce the biomass for more acclimatization. After a certain number of cycles, it has been observed that level of sludge in serum bottles has increased which shows there was growth of adapted sludge. The temperature and pH values measured during the cycles in reactors were "28±1°C" and initial pH of "7.6±0.1" and at the end of cycles the recorded pH was "6.3±0.1". This drop in pH could be because of volatile acid formation in anoxic conditions and these acids are being utilized by SRB to reduce sulphates to sulfides. The percentage of color removal in the reactors at the end of each cycle is presented in Fig. 1. From the results, it is clear that on acclimatization of the sludge in sulfidogenic environment decolorization of the dye improved and it reached from 69% in 7 days to 84% in 2 days cycle time. These results prove that decolorization of Tartrazine can be achieved in sulphidogenic environment easily and has an application potential. There are probably at least two mechanisms for the decoloration of azo dyes in bacterial systems: (i) direct electron transfer to azo dyes as terminal electron acceptors via enzymes during bacterial catabolism, connected to ATP-generation (energy conservation), and (ii) a gratuitous reduction of azo dyes by the end products of bacterial catabolism, not linked to ATP-generation; in this case it is the H_2S formed as a result of sulphidogenesis .

Fig. 2 shows the effect of initial pH in the decolorization of dye. The results show that optimal pH for decolorization is between 8 and 9 and effective color removal (87-92%) could be achieved in a day. It is also seen that alkaline pH is more favoured than acidic pH for decolorization of Tartrazine. The results show that the decolorisation took long hours if the pH is not maintained in the optimal range. Chang et al. [11] found that the dye reduction rate increased nearly 2.5-fold as the pH was raised from 5.0 to 7.0, while the rate became insensitive to pH in the range of 7.0–9.5. Similar results are obtained in this study.

B. COD oxidation and Sulphate reduction

The percentage removal of COD after the end of each cycle in the reactors during the feasibility study is given in Fig. 3. In each cycle COD/sulphate ratio is maintained at one with each approximately equal to 2500 mg/l. The removal of COD has increased after each cycle and reached almost 73% at the end of fifth cycle. The electrons released from electron donor are transferred to azo dye and sulphate, resulting in color removal, sulphate reduction, and, therefore, decreased COD amount.

The percentage removal of COD in SBRs used for studying the effect of pH is shown in Fig.4. The results show that the highest reduction of COD was 81% when the initial pH was adjusted to 8.0. Also, it is to be noted that the trend of COD removal is in agreement with the percentage removal of color, indicating that there is a mineralization of the dye as a result of sulphidogenesis process. The control reactor with initial pH (7.6±0.1) showed only 66% removal of COD and took more time to degrade compared to more alkaline pHs 8 and 9. Therefore, the results support that alkaline pHs remove more color and COD.

Fig. 5 shows the percentage removal of sulphates in SBRs used for studying the effect of pH. The results show that in alkaline pHs the percentage of sulphate removal is efficient and highest removal of 79% was obtained at pH 9. From the trends in removals of color (Fig.2), COD (Fig.4) and Sulphate (Fig.5), it is evident that the decolorization of Tartrazine associated with the sulphidogenesis process.

C. Sulphide production

As the bacteria enriched in the reactor are sulphate reducing type, the sulphate is being converted to sulfide; this was evident from the rotten egg smell sensed while opening the serum bottles at the end of each cycle for analysis of the parameters. This is due to more percentage sulfides as free H_2S due to the acidic pH (6.3±0.1). At this acidic pH, 90% of the sulfide is in gaseous form, which tends to escape on opening the reactors, while the remaining is in a soluble form that was analysed and in the range of 150-220 mg/l.

D. ORP measurements

At the end of each cycle, ORP in the serum bottles was measured. The values were always

found to be in the range of "-280 to -400 mV", which governs the anaerobic/anoxic condition in the reactors conducive for sulphidogenesis.

I. FIGURES

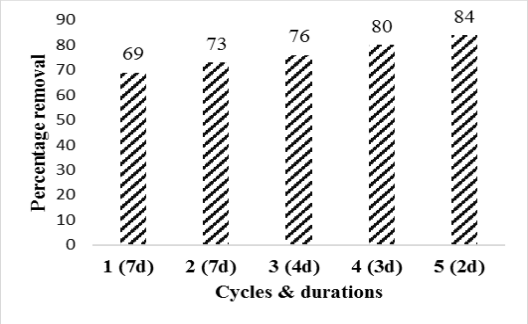

Fig.1 Percentage color removal after each cycle

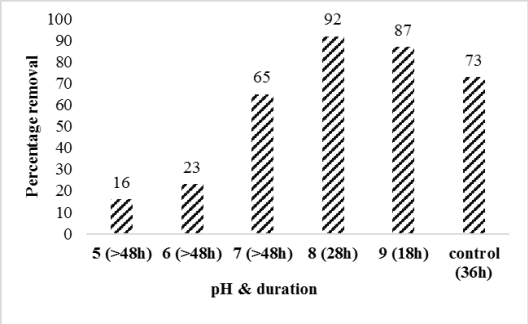

Fig.2 Percentage color removal at each pH

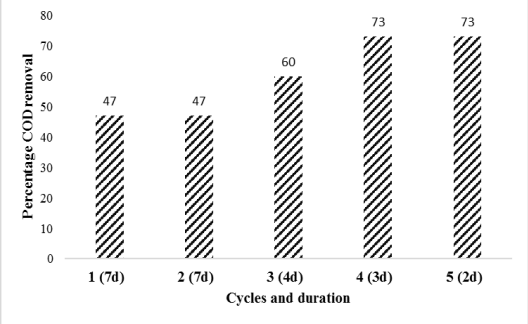

Fig. 3 Percentage removal of COD after each cycle

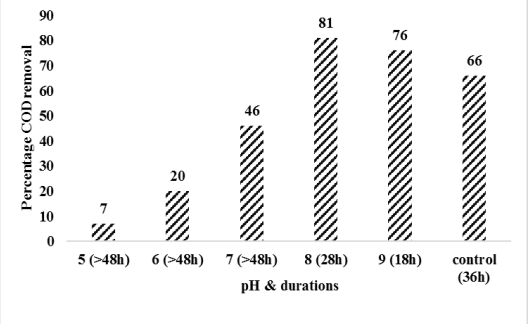

Fig. 4 Percentage removal of COD at each pH

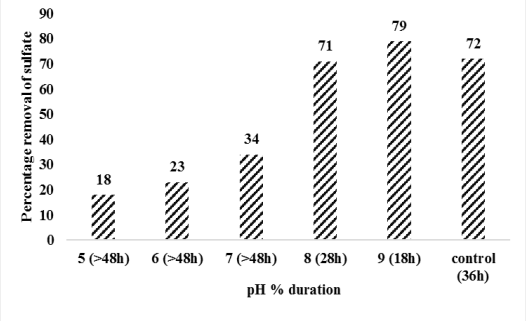

Fig. 5 Percentage removal of sulphate at each pH

V. Conclusion

The results of this study show that activated sludge could be used as an effective seed sludge for developing sulphidogenesis process for decolorization of Tartrazine dye at 100 mg/l. The effective dye removal could be achieved in SBR operation in 2 days on acclimatization of biomass. The time for decolorization could be reduced to around 24 hours at an optimal pH between 8 and 9. The results of this study indicate an economical and ecofriendly technique for decolorization of Tartrazine at 100 mg/l.

VI. References

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