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ELECTRODE COMPOSITION OF REACTIVE BLACK 5 DYE IN AQUEOUS SOLUTION

HASSAN M. TAMOUS

*Assistant professor of physical Chemistry [Electrochemistry] Al Azhar University-Gaza,
Faculty of Science, Department of Chemistry, Gaza Strip, Palestine.*

ABSTRACT

High oxygen evolution overpotential and inexpensive $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ electrodes were investigated in this work. These electrodes were prepared and used as anodes for removal of Reactive Black 5 (RB5) dye in aqueous solution. In some local cases in Palestine, wastewater color was found to be the only problem in meeting local effluent standards. This investigation reveals that both electrodes can remove the color of these dyes completely. After 10 min, nearly complete degradation of RB5 was achieved (99.2%, and 91.90) using $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ electrodes respectively at pH 6.8, current density 30 mA cm^{-2} and in the presence of NaCl (4 g L^{-1}) at 25°C . At the same condition the optimal time for COD removal for $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ electrodes was 7 and 9 hours.

KEY WORDS: Removal, reactive black 5, electrodes, electrodeposition, electrocatalytic oxidation.



HASSAN M. TAMOUS

Assistant professor of physical Chemistry [Electrochemistry]
Al Azhar University-Gaza, Faculty of Science,
Department of Chemistry, Gaza Strip, Palestine.
Dr.hassantamous@gmail.com

INTRODUCTION

Several recent studies report the use of electrooxidation to treat model aqueous solutions containing various dyes. Rajkumar et al. [1] studied the electrochemical degradation of Reactive Blue 19 over a titanium-based dimensionally stable anode regarding the effect of operating conditions (current density, salinity, reaction temperature and initial dye concentration) on treatment performance, while they also identified major reaction intermediates. The effect of various operating conditions on Acid Blue and Basic Brown degradation over a lead/lead oxide anode and on Acid Orange 7 degradation over a boron-doped diamond anode was studied by Awad and Abo Galwa [2] and Fernandes et al. [3], respectively. Basic Yellow 28 and Reactive Black 5 were used as test substances to compare the efficiency of a diamond electrode to that of conventional metallic electrodes (iron, aluminium and copper) [4], while an activated carbon fiber electrode was used to assess the electrochemical degradability of 29 different textile dyes [5]. In further studies [6], several advanced oxidation processes, namely wet oxidation, TiO₂ photocatalysis, electro-Fenton and UV-assisted electro-Fenton were compared concerning their efficiency in treating Reactive Black 5. The removal of the color from synthetic wastewater containing Reactive Black 5 was experimentally investigated using direct current electrocoagulation at iron electrodes [7], Fenton-like reaction [8], nano zerovalent iron (NZVI) [9], Fe/Cu co-doped TiO₂ nano-particles under mercury lamp [10], and over a titanium-tantalum-platinum-iridium anode [11]. The effects of operational parameters such as current density, initial pH, electrolysis time, NZVI dosage, UV light, catalyst, and Na salt, initial dye concentration and solution conductivity on color removal efficiency were investigated.

Vlyssides et al. [12] investigated the electrooxidation of textile effluents over a Ti-Pt electrode at different chloride concentrations and reported that electrochemical treatment improved the biotreatability (as assessed by the BOD/COD ratio) of the original effluent.

Naumczyk et al. [13] compared the decolorization and mineralization rates of textile effluent electrochemical degradation over three Ti-based electrodes coated with different metals and they also attempted to identify major reaction by-products. Sakalis et al. [14] demonstrated a continuous, pilot-scale cascade electrochemical reactor capable of achieving 90% decolorization of a textile effluent at a residence time of 40 min. The mechanism electrodegradation of Reactive Black 5 (RB5) dye may be occurred in electrocatalytic oxidation [3, 15] as well as the electrochlorination method [16]. The purpose of this work, is to remove the Reactive Black 5 (RB5) dye electrochemically using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and (Ti/(SnO₂+Sb₂O₃)) electrodes. Different factors including the pH, concentration of electrolyte, conductive electrolyte type, current density, time of electrolysis, initial concentration of RB5 solution, and temperature were studied and optimized.

EXPERIMENTS

Chemicals and Instrumentation

Sodium chloride, sodium fluoride, sodium carbonate, sodium sulphate, calcium chloride, potassium chloride, sodium hydroxide, sulphuric acid, potassium dichromate, silver sulfate, sodium thiosulphate, nitric acid, sodium fluoride, tin tetrachloride pentahydrated, antimony trichloride, 2-propanol, oxalic acid, cobalt nitrate hexahydrate and lead nitrate were of analytical grade and purchased from Merck. Reactive Black 5 dye was purchased from Dr. Ehrenstorfer GmbH (Germany) with purity of 99.9%. Other reagents were of the analytical grade. Distilled water was used for the preparation of solutions. Standard solutions of potassium dichromate (K₂Cr₂O₇), sulfuric acid (H₂SO₄) reagent with silver sulfate (Ag₂SO₄) were prepared to measure the COD. Different standard solutions of Reactive Black 5 dye with concentration from 10 – 70 mg/L were prepared to measure its degradation at different conditions. The double-beam UV-visible

spectrophotometer is from Shimadzu, the DC power supply is model GP4303D, LG Precision CO. Ltd. (Korea), a pH meter model AC28, TOA electronics Ltd., (Japan) to adjust pH of the solutions and a digital multi-meter is kyoritsu model 1008, (Japan) for reading out the current and potential values. A closed reflux titrimetric unit was used for the COD determination.

Preparation of Ti/(SnO₂+ Sb₂O₃) and Ti/Co₃O₄/PbO₂/(SnO₂+ Sb₂O₃) electrodes

Treatment of titanium surface

Pretreatment of titanium surface was carried out according to the procedures suggested by Yang et al [17] as follows: The titanium sheet was polished on 320-grit paper strips, using water as lubricant, followed by 1Mm siliceous past blasting. The sheet was then degreased in 40% NaOH, cleaned in a hot 1:1 mixture of HNO₃ and H₂SO₄ and finally washed in distilled water. The treated surface was immersed in a boiling aqueous solution of oxalic acid (15%) until the TiO₂ dissolved. Coating by active metal oxide was carried out immediately to minimize formation of TiO₂ [18].

Coating Co₃O₄ on Ti-surface

Co₃O₄ electrodes were prepared by thermal decomposition of nitrate precursor deposited onto a titanium plat. Immediately after pretreatment, a 0.5 M Co(NO₃)₂ x 6H₂O in isopropanol solution was layered onto the support. Then the support was dried at 60⁰C to evaporate the solvent and heated in air furnace at 300 °C for 10 min. Oxide layers were deposited by repeating this procedure 10 times to achieve full coverage of the metallic surface. Final annealing was done by keeping the sample for one hour at 300⁰ C to complete the thermal decomposition [19].

Electrodeposition of PbO₂ layer on Ti/Co₃O₄-surface

The electrodeposition of PbO₂ was performed at constant anodic current (100mA, 30 min) in 0.1 M HNO₃ solution containing 0.5 M Pb(NO₃)₂ and 0.04 M NaF. During electrolysis the potential was ranged from 1.5 to 1.8 V. Electrodeposition was

carried out for 30 min, achieving a PbO₂ loading of about 14 mg cm⁻² (by weighting the electrode before and after the coating). In order to verify the reproducibility of the electrode preparation, experimental runs were repeated with three different electrodes prepared according to the procedure described above. Results showed a fair reproducibility: current efficiencies measured on the three electrodes (when the same operative conditions were adopted) differed by about 5%. Electrodes were also sufficiently stable to allow their use in consecutive runs: the analyses of solutions revealed that no metal dissolution occurred during electrolyses [18].

Coating of mixture of SnO₂ and Sb₂O₃ on Ti/Co₃O₄-surface

The thermal deposition of SnO₂ on titanium sheet was carried out according to the procedure of Lipp and Pletcher [20] as follows: The pretreated titanium sheet was immersed for some minutes in a solution of 20% wt SnCl₄.5H₂O and 2% wt SbCl₃ in 2-propanol. The excess of alcohol was allowed to evaporate by heating in air at 90⁰C for 10 min. This stage was then repeated. After two applications, the oxide layer was formed thermally by heating at 500 °C for 20 min in a muffle furnace, with a low and continuous flow of oxygen. These stages were repeated until the SnO₂ coating reached a loading of 0.8 (± 0.01) mg cm⁻² it was normal to employ five applications. Finally, the coating was annealed at 500⁰C for 60 minutes [21].

Electrolysis of Reactive Black 5 Degradation

Galvanostatic electrolyses were carried out at Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and (Ti/(SnO₂+Sb₂O₃)) electrodes, with current density ranging from 0 to 200 mAcm⁻² and electrical potential ranging from 1-12 volts. Runs were performed at 10 - 40⁰C. Solutions of 100 mg L⁻¹ of pure RB5 solution were used. The investigations of this study were carried out in the presence of sodium chloride (0.2-20 g L⁻¹) and 4 g L⁻¹ of different conductive electrolytes such as; NaCl, CaCl₂, KCl, Na₂CO₃, NaF, NaPO₄ and Na₂SO₄ with pH between 1.5 and 12. The electrolysis duration ranges from 0-30 min. The electrochemical degradation of the RB 5

solutions was carried out in a 100 mL Pyrex glass cell where the prepared electrodes $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ work as anode and austenitic stainless steel as cathode. The electrodes were connected to a DC power supply while the current and potential measurements were read out using digital multi-meter.

Analysis

Two main parameters were measured to evaluate the electrochemical treatment efficiency, the remaining pollutant concentration and the COD. Remaining pollutants (RB5) concentration was measured with the double-beam UV-visible spectrophotometer from Shimadzu at $\lambda_{\text{max}}=595$ nm using calibration curve with standard error ± 0.5 [22]. The COD was determined using a closed reflux titrimetric method [23].

RESULTS AND DISCUSSION

Effect of Various Factors on the Rate of Degradation

Effect of pH value

The pH of the solution was varied while the other conditions were kept constant. As shown in Fig. 1, maximum removal of RB5 was achieved at pH 6.8 for $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ respectively. The pH values of the solutions were adjusted by adding drops of H_2SO_4 and NaOH . The reactions were carried out for 10 min for two electrodes under the following conditions: the initial concentration of 100 mg L^{-1} , a current density of 30 mA cm^{-2} , a temperature of 25°C and NaCl concentration of 4 g L^{-1} . The distance between the two electrodes was adjusted to 1cm. It was found that the maximum rate of degradation using $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ electrodes was achieved in neutral medium as the optimal medium.

Effect of the NaCl concentration

Different concentrations of NaCl were applied to study their effect on the removal of RB5 as indicated in Fig. 2. The results indicate that an increase of the electrolyte concentration up to 4

g L^{-1} lead to increase in the RB5 degradation rate for two $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ electrodes. The NaCl solution liberates Cl_2 gas which is considered as the active species for the degradation of organic compound. Further increase of the NaCl concentration has slightly effect on the degradation rate of RB5.

Effect of current density

As shown in Fig. 3, RB5 removal increase with increasing the applied current density up to 30 mA cm^{-2} by using $\text{Ti}/\text{Co}_3\text{O}_4/\text{PbO}_2/(\text{SnO}_2+\text{Sb}_2\text{O}_3)$ and $(\text{Ti}/(\text{SnO}_2+\text{Sb}_2\text{O}_3))$ electrodes. Further increase of the current density was followed by a gradual decrease in RB5 degradation due to increase in temperature. Above a temperature 35°C , sodium hypochlorite tends to chemically decompose to sodium chlorate (8).



(8)

So when temperature rises higher than 35°C , production of NaClO falls. The rate of hypochlorite decomposition increases with increase in current density.

Effect of type of electrolyte

Electrolytes of 4 g L^{-1} of the following salts; NaCl , CaCl_2 , KCl , Na_2CO_3 , NaF , Na_3PO_4 and Na_2SO_4 were studied by three electrodes. As appears in Fig. 4, The NaCl , KCl and CaCl_2 were the most effective conductive electrolytes for the electrocatalytic degradation of the investigated RB5 while NaF and Na_2SO_4 electrolytes show poor results. The Cl^- anion is a powerful oxidizing agent. These observations were also confirmed in other studies [23-25].

Effect of temperature

It is well known that the rate of diffusion of ions increases with increasing temperature. Fig. 5 represents the correlation between the concentration of the remaining RB5 dye as a function of the solution temperature. The rate of the RB5 degradation increase significantly with increasing the solution temperature until 25 or 30

°C. Therefore, 25 °C was fixed as optimal electrolysis temperature under the same conditions mentioned respectively.

Effect of initial RB5 concentration

Fig. 6 shows the effect of different initial RB5 concentrations on the rate of RB5 degradation. Removal of the RB5 can be achieved in the presence of initial RB5 load up to 100 mg L⁻¹. However, increasing the RB5 concentration above this level results in a decrease in the electro-catalytic rate of degradation. The removal efficiency of the RB5 by using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes at 100 mg L⁻¹ was the optimum concentration for the initial load concentration of RB5. As the initial RB5 concentration increase, the degradation efficiency decrease. This evidence that the generation of the powerful oxidizing agent Cl⁻ ions on electrode surface was not increased in constant current density.

Effect of distance between the cathode and anode

The effect of distance between the two electrodes of the cell was studied. It was found from Fig. 7 that there was an increase of hypochlorite generation by decreasing the distance between the two electrodes up to 1cm for Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes. Therefore 1 cm was chosen as optimum distance between electrodes for sodium hypochlorite generation. The experiments were carried out under the following conditions; current density 30 mA cm⁻², pH of 6.8, temperature of 25 °C and the concentration of NaCl 4 g L⁻¹. The time of electrolysis was 10 min. It is clear that the sodium hypochlorite production increase with decreasing distance down to 1cm. This is due to drop of electrolyte ohmic potential, and hence the cell voltage [26]. The highest hypochlorite production was achieved with narrow distance between the cell electrodes of 1cm.

Effect of the electrolysis time

To assess the effect of electrolysis time, experiments were conducted with operating

treatment conditions that were consistent with those described for Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes. The maximum removal of RB5 was achieved using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes after at least 10 min. Therefore, this was taken as optimal degradation time for the removal of RB5. The optimal time for COD removal for Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes was 7 and 9 hours as shown in Fig.8.

Comparison

At optimized conditions, the percentages of RB5 degradation for Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes are 99.2% and 91.90%. The results indicate that the Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) electrode is more adequate than Ti/(SnO₂+Sb₂O₃) electrode for the degradation of RB5. These behaviors may be attributed to the presence of PbO₂ which cause the electrode more high oxygen evolution overpotential. The percentages of degradation for each method using in literature and the electrochemical method in this work were represented in the Table 1. It is clear that the electrochemical degradation the best.

Application of the treatment process in real wastewater samples

The treatment of RB5 effluents obtained from dyeing factories was carried out by using the prepared Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) modified electrodes. The treatment was performed, first by collecting actual waste samples from the wastewater effluents of the RB5 dyeing bath. The initial dye load concentration of these samples were 150 mg/L taken from Hubbub dyeing factory located in the industrial area at Biet Hanon, Gaza Strip, PNA. The dyestuff solutions were treated by the electrocatalytic oxidation technique by the same method as applied to the treatment of RB5 in aqueous solution to investigate the optimum condition for real wastewater containing the dye. After the treatment process, the removal percent of RB5 at 10 min using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and

Ti/(SnO₂+Sb₂O₃) electrodes was (98.5% and 89.1%) respectively, and removal percent of COD showed that (100% at 8 and 10 hr). These results indicate that the suggested modified

electrodes are highly efficient in the treatment of effluents containing RB5 dye with very slight effect of matrix.

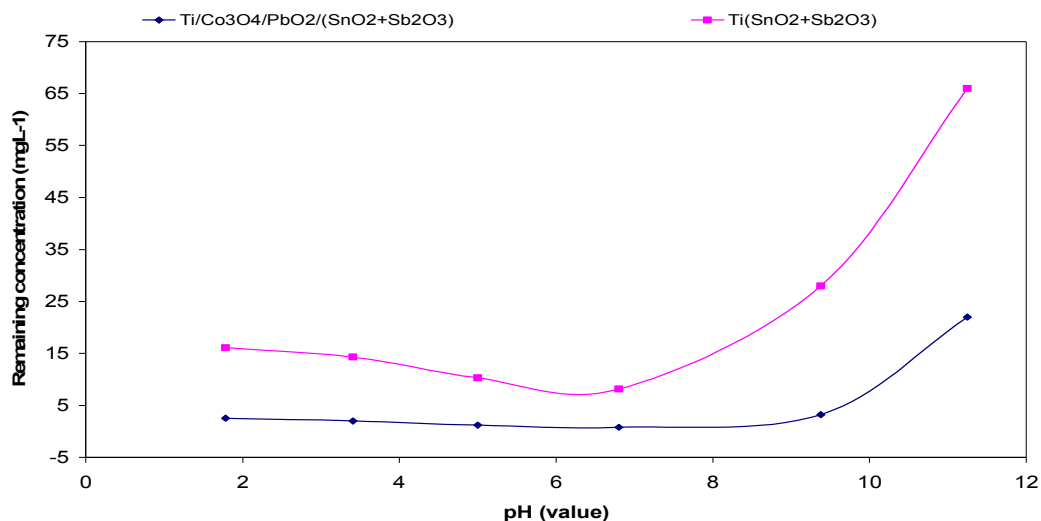


Fig. 1

Figure 1
The effect of pH on RB 5 using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes.

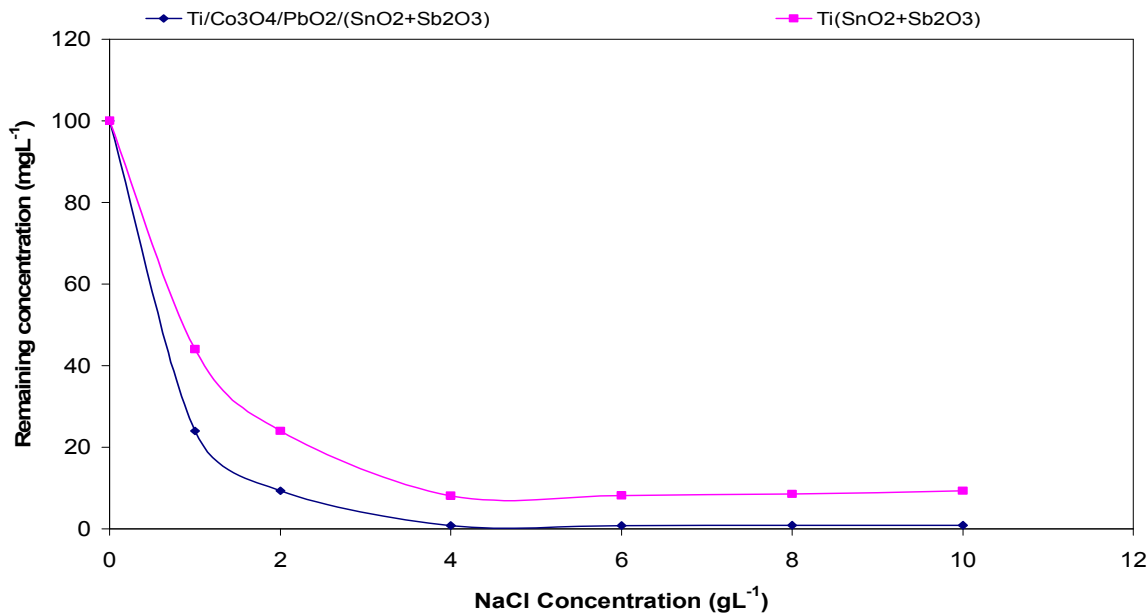


Fig.2

Figure 2
The effect of NaCl concentration on RB 5 using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) And Ti/(SnO₂+Sb₂O₃) electrodes.

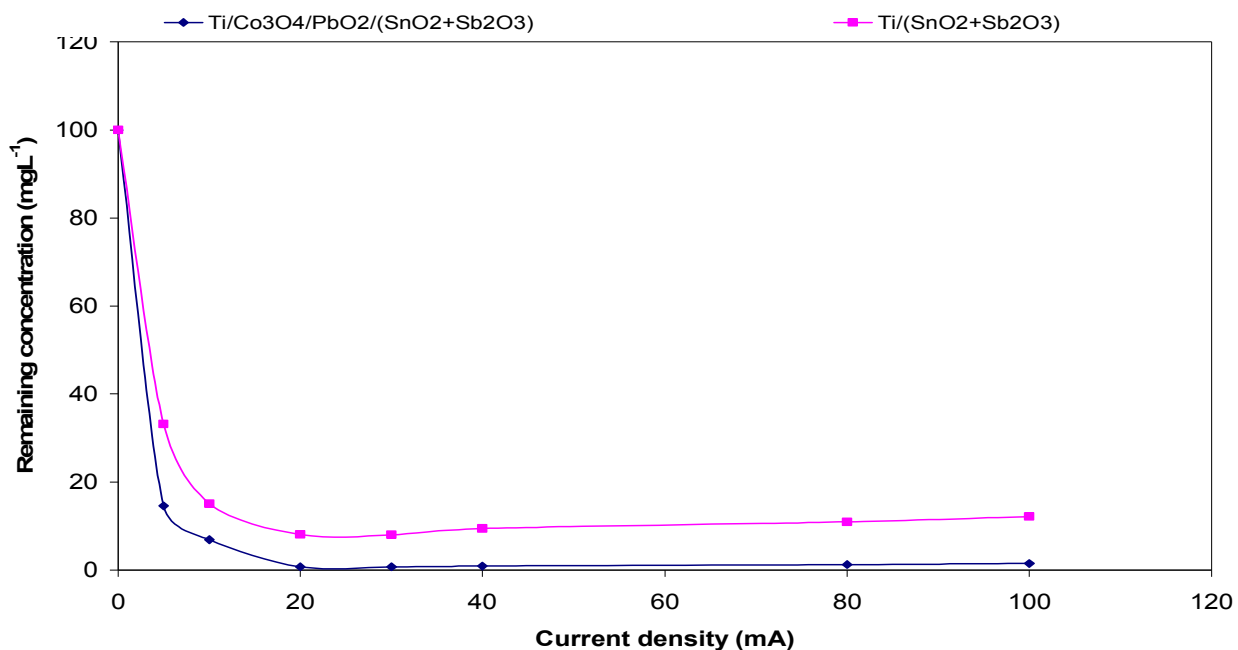


Fig.3

Figure 3
The effect of current density on RB 5 using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) And Ti/(SnO₂+Sb₂O₃) electrodes.

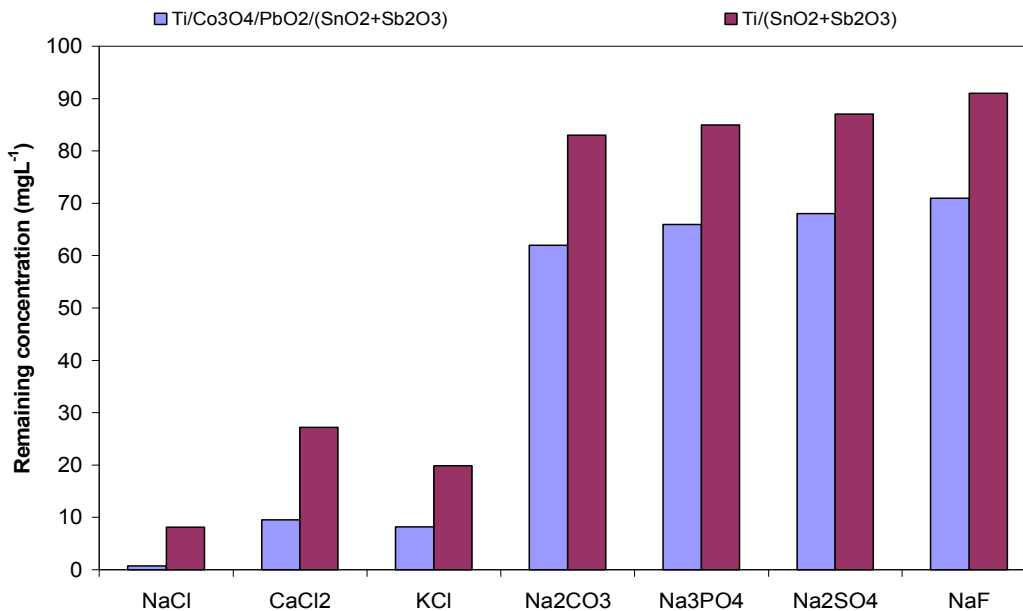


Fig.4

Figure 4
The effect of the conductive electrolyte type on RB 5 using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes.

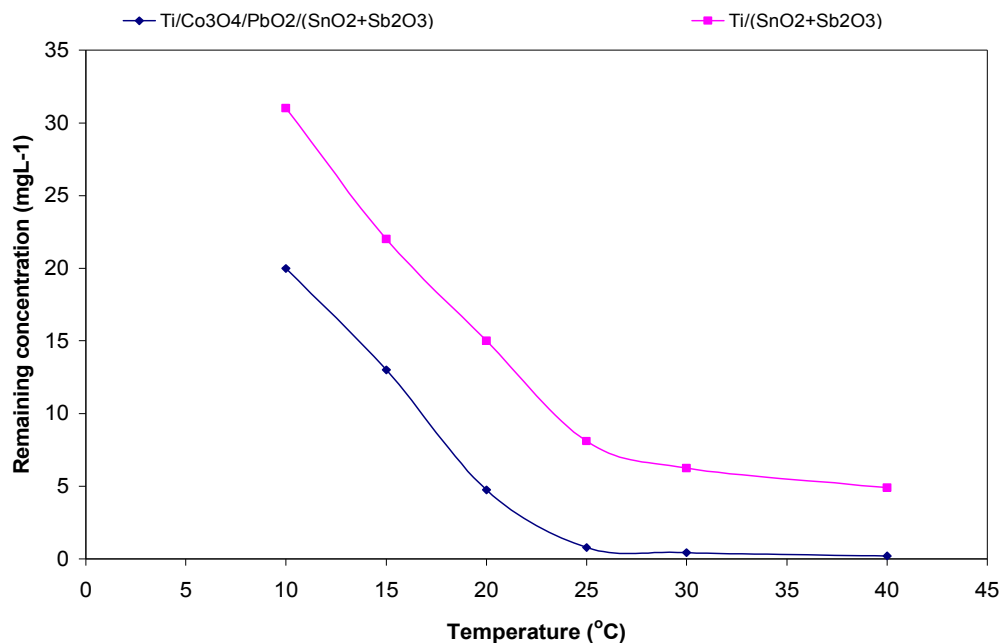


Fig.5

Figure 5
The effect of temperature on RB 5 using $Ti/Co_3O_4/PbO_2/(SnO_2+Sb_2O_3)$ And $Ti/(SnO_2+Sb_2O_3)$ electrodes.

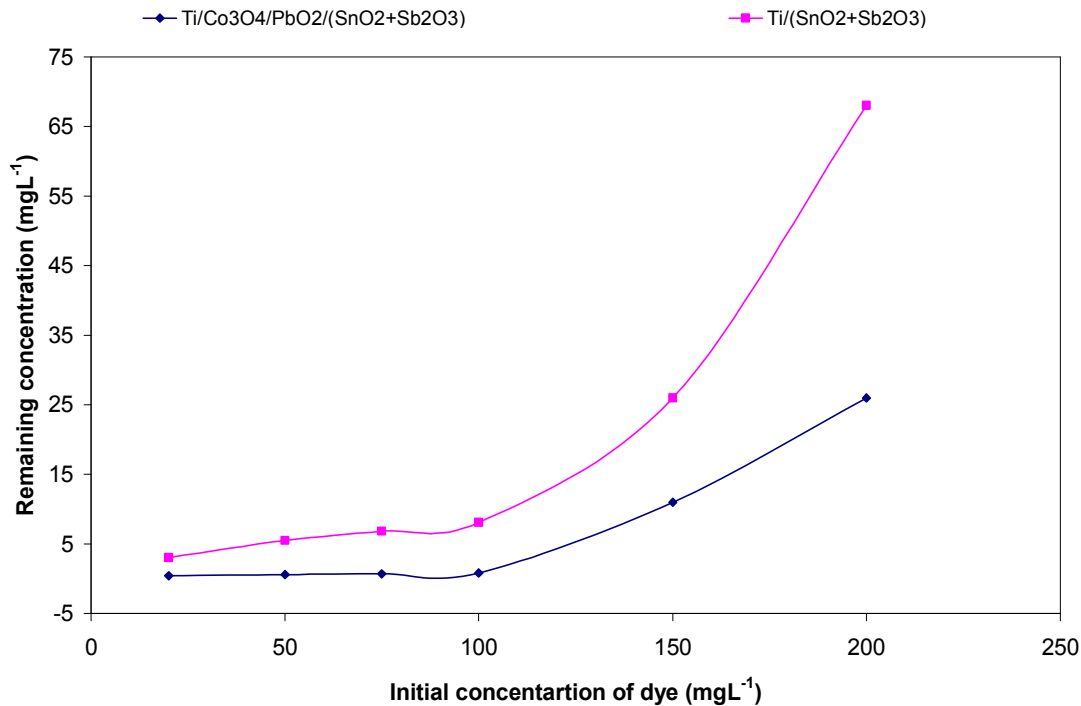


Fig.6

Figure 6
The effect of initial concentration on RB 5 using $Ti/Co_3O_4/PbO_2/(SnO_2+Sb_2O_3)$ And $Ti/(SnO_2+Sb_2O_3)$ electrodes.

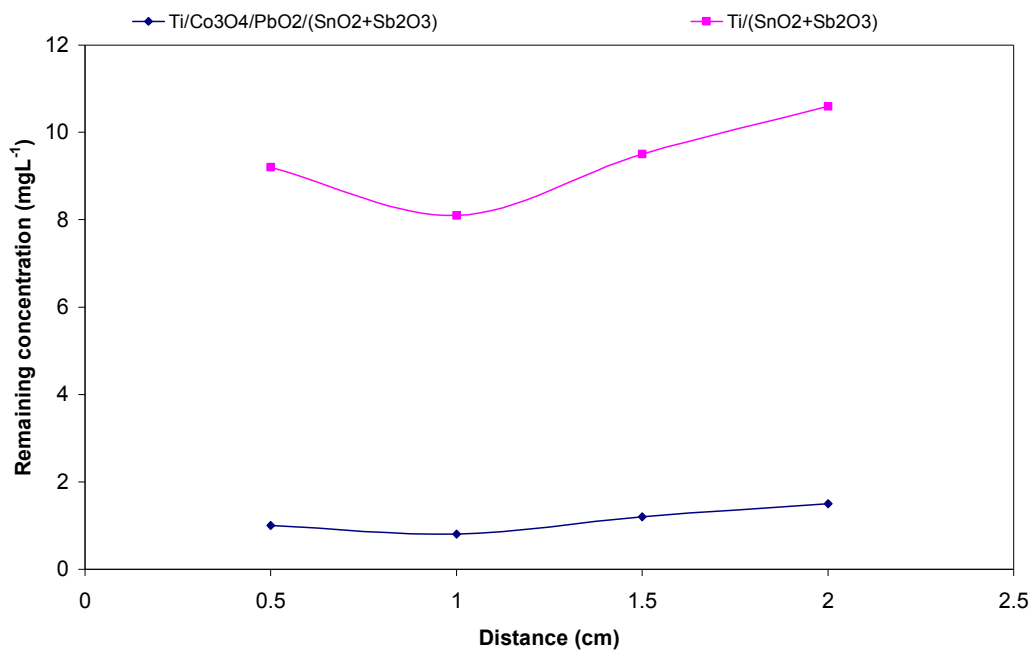


Fig.7

Figure 7

The effect of distance between the cathode and anode on RB 5 using *Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃)* and *Ti/(SnO₂+Sb₂O₃)* electrodes.

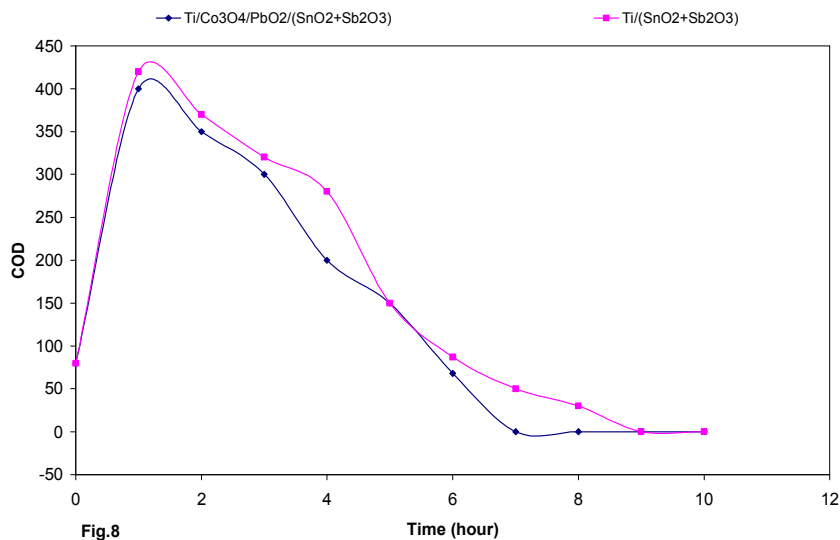


Fig.8

Figure 8

The effect of time on COD removal using *Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃)* and *Ti/(SnO₂+Sb₂O₃)* electrodes.

Table 1
Comparison of proposed electrodes and some decolonization technologies For Reactive Black 5 (RB5) dye removal

Method of treatment	Initial dye concentration (mg L ⁻¹)	Color removal efficiency (%)	Operating time	Literature
Fenton	100	99	20dk	27
UV/TiO ₂	100	80	30 min	28
Electro-Fenton	100	40	30 min	28
UV	100	79.7	240 min	29
UV/Fe ₂	100	92.6	240 min	29
Photo assisted Fenton	54	70	480 min	30
Anaerobic mixed culture	3200	98	30 h	31
<i>C. oleophila</i> yeast	200	95	24 h	32
Ozone assisted electrocoagulation	100	94	30 min	33
Ti/Co ₃ O ₄ /PbO ₂ /(SnO ₂ +Sb ₂ O ₃)	100	99.2	10min	This work
Ti/(SnO ₂ +Sb ₂ O ₃) electrode	100	91.9	10min	

CONCLUSION

In this Study the Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes were prepared by electrodeposition and used as anodes for electrodegradation of RB5 in aqueous solution at different parameters. The optimum condition for two electrodes are NaCl (4 g L⁻¹), temperature at 25°C, degradation time of 10 min, initial concentration of 100 mg L⁻¹, current density

equals 30 mA cm⁻² and 1cm distance between the two electrodes of the cell. The degradation of RB5 was nearly completed (99.2 % and 91.9%) using Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes at pH 6.8, respectively. The optimal time for COD removal for Ti/Co₃O₄/PbO₂/(SnO₂+Sb₂O₃) and Ti/(SnO₂+Sb₂O₃) electrodes was 7 and 9 hours

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