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A Kinetic Investigation of Electrochemical Degradation of 2-N, Ndimethyl-4-aminophenyl azobenzene carboxylic acid dye at Zr/graphite Modified Electrode in Aqueous Solution

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ABSTRACT

The electrochemical degradation of wastewater containing methyl red dye was investigated by synthesized Zr/graphite modified electrode. The kinetics of degradation by graphite and Zr/graphite modified electrode are compared. Methyl red is one of the most important textile dyes. The wastewater of methyl red can be oxidized to anthranilic acid, which is hydrolyzed to N, N–Dimethyl-p phenyldiamine a compound that is toxic to aquatic life. SEM was observed for synthesized Zr/ graphite modified electrode. The thin film formation of Zr or encapsulated in graphite rod is observed from SEM/EDX. The UV-Visible spectra for before and after degradation were studied for methyl red dye. The effect of concentration of the dye and the current on the kinetics of degradation was studied. The ICE values of different experimental conditions are evaluated. The anodic oxidation by Zr/graphite modified electrode showed the complete degradation of water containing methyl red, which is confirmed by UV-Visible and COD measurements. The dye is converted intoCO₂, H₂O and simpler inorganic salts. The results observed for reuse of modified electrodes indicates that the Zr/graphite modified electrode would be promising anode for electrochemical degradation of methyl red. This method can be applied for the remediation of waste water containing organics, cost effective and simple.

Graphical Abstract



Energy -dispersive X-ray spectroscopy (EDX) for Zr/graphite modified electrode.

Keywords: Methyl Red, Anodic oxidation, Zr/graphite modified electrode, Mineralization.

INTRODUCTION

The graphite electrode (GE), as a type of carbon electrode, has attracted considerable interest because of its low cost, avail-ability, and easy of modification, which ensure good characteristics comparable to other carbon materials, such as carbon paste and glassy carbon electrodes [1-4].Graphite is described as two-dimensional crystal, consist of a single layer of carbon atoms. It is interesting for hypothetical as well as experimental researchers in the universe. Graphite contains numerous Graphene-sheets held collectively by weak Vander Waals forces. Thus accredit by a single layer of carbon atoms thickly enclosed in a comb like complex [5-7].Graphite has both metallic and nonmetallic properties being thus useful as electrode material. According to Chehreh Chelgani et al. 4% of the world graphite is used to produce pencils consisting of fine graphite powder in an inorganic (resin) or organic matrix (clay or a high polymer, e.g., cellulose) [8-10]. Electrochemical degradation of organic pollutants has been investigated by many researchers, and it has been proved as a costeffective technology. Thus electrochemical degradation is a good choice of removing MR. As a new and attractive technology, electrochemical oxidation of organic pollutants has gained significant attention over the past two decades [11, 12]. Methyl Red (MR) is a commonly used mono azo dye in laboratory purpose, textiles and other commercial products. It was well known that MR dye has been used in paper printing and textile dyeing [13]. Azo dyes contain one or more nitrogen to nitrogen double bond (-N-N-) and they constitute almost 80% of the reactive dyes. Most of the azo dyes are not biodegradable by aerobic treatment processes. Advanced oxidation processes involving Fenton's and photo-Fenton's process for the degradation of organic dyes are extensively reported in the literature [14]. Under the action of current, some of the organic matter in the wastewater may decompose into low molecular organic matter, further oxidized to oxides of C, N, S and H₂O [15, 16]. In the present work the electrochemical method has been developed for the electrochemical degradation of Methyl Red (MR) dye with Zr/graphite modified electrode.

MATERIALS AND METHODS

Methyl Red (E-Merck) solution was prepared [0.0001N]. The 2.5 cm length and 0.63 cm diameter graphite electrode (ALFA AESAR) was used. A graphite electrode is dipped into the above solution. Prior to experiment the surface pre-treatment of graphite electrode was performed by hand polishing of the electrode surface with successive grades of emery papers down to 3000 grit up to a mirror finish. The polished electrode was then degreased with 1:1 HCL solution, acetone and washed with running double distilled water and utilized for further electro deposition. The experimental set up is shown in the figure 1. Zirconium was deposited on graphite electrode from ZrCl₃ (Arora matthey) solution. All chemicals were used of accepted grades of purity. The experimental apparatus is shown in figure 2. It consists of reaction chamber and a voltage power supply. The electrode system consists of a graphite modified electrode as anode and Pt electrode as cathode in case of degradation of dye by anodic oxidation with graphite electrode. The distance of the anode and cathode was 2cm. The kinetic runs were carried out with graphite electrode alone as anode. In another case, Zr(III) thin film deposited on a conductive Graphite electrode has taken as anode and Pt electrode as cathode. The experiment was run from 6 to 660minutes with continuous stirring. The kinetic runs were carried out at different concentration of methyl red with different applied current. To account for the mineralization of the dye solution COD was measured at regular intervals of time. The pH of the solution before and after degradation was measured. A positive voltage of [12V-20V] was applied by using battery eliminator (NEULITE INDIA) and current output of 2.0 mA-6.0mA using rheostat (INSIF INDIA). The decolorization and disappearance of methyl red was followed by using spectrophotometer (ELICO SL 171).



Figure 1. Synthesis of Zr/GME by electrochemical method.

Table 1. Properties of Methyl Red Dye





Figure 2. Experimental set up for electrochemical degradation of MR Dye.

RESULTS AND DISCUSSION

Reaction with Graphite as anode electrode

Effect of Methyl Red: The experiment were performed by taking at different concentration of Methyl Red dye solution (E-Merck) varying $(0.5 \times 10^{-4} \text{ to } 2.0 \times 10^{-4} \text{ N})$ and keeping constant current (4.0 mA). The change in concentration of the MR was recorded by change in color using spectrophotometer. This indicates with the increase of concentration of dye the rate of the reaction is decreased. Surface of the graphite electrode, decreases the rate of flow of current across the electrode-solution interface, which decreases the concentration of OH radicals. A plot of log% T (percentage transmittance of light) versus time in min was linear up to 60% of the reaction indicating disappearance of MR follows 1st order kinetics. The rate constant values are given in table 2 and figure 3(a), the pH value before and after the degradation remain constant. The COD values for MR solution before and after degradation

were measured and are reported in table 2, figure 3 (b). The decrease of COD values to $<50 \text{ mg L}^{-1}$ after degradation indicates >98% degradation of MR.

Table 2. Effect of Concentration of MR on the rate of degradation and COL	values
[Current = 4.0mA, Temperature = 298K]	

10-4	10 ⁴ k	Effect	of pH	COD Value	es in mg L ⁻¹
[MR] In N	in Sec ⁻¹	Before degradation	After degradation	Before degradation	After degradation
0.5	0.537	6.15	6.08	416	48
1.0	0.460	6.19	6.05	480	44
1.5	0.268	5.99	5.91	576	46
2.0	0.191	5.57	5.32	608	48



Figure 3. [A] Effect of Concentration of MR on rate of degradation and [B] COD Values.

Effect of current on the rate: To investigate the effect of applied current the experiments were carried out at five different currents varying from 2.0 mA - 5.0 mA and keeping MR dye as constant .From current study, it is clear that with increase of the current the concentration of oxidizing intermediates and OH radicals increases. This increases the rate of reaction and hence increases the rate of degradation. Rate constant values are reported in table 3 and figure 4 (A). The COD values for different current of MR solution before and after degradation were measured and are reported in table 3, figure 4 (B).

Table 3. Effect of current on the rate of degradation and COD values $[MR = 1.0X10^{-4} N, Temperature = 298K]$

ſ	Current in	10 ⁴ k	COD Value	es in mg L ⁻¹		
	mA	In Sec ⁻¹	Before degradation	After degradation		
Γ	2.0	0.076	480	48		
I	3.0	0.191	480	42		
I	4.0	0.460	480	44		
	5.0	0.537	480	48		



Figure 4. [A] Effect of current of MR on the rate of degradation and [B] COD Values.

Effect of temperature: To investigate the effect of temperature the experiments are carried out at three different temperatures, the increase of temperature the rate of the reaction is increased. It is observed that the rate of removal of colour is not very significant at low temperature. However the reaction is more significantly influenced at higher temperature, the rate constant and COD for before and after degradation of dye is reported in table 4 and figure 5(A). Since the diffusion rate is increased with temperature, an increase of temperature could bring about an increase in the degradation rate. Thermodynamic parameters are calculated and are reported in table 5.

Table 4. Effect of Temperature on the rate of degradation and COD values $[MR=1.0X10^{-4} N, Current = 4.0mA]$

Tomporatura	10 ⁴ k	COD Value	es in mg L ⁻¹
in K	in Sec ⁻¹	Before degradation	After degradation
293	0.422	480	32
298	0.460	480	44
303	1.343	480	33



Figure 5. [A] Effect of temperature of MR on the rate of degradation and [B] COD Values.

Temperature in K	$\Delta \mathbf{H}^{\#}$ KJ mol ⁻¹	∆S [#] J K ⁻¹ mol ⁻¹	$\Delta \mathbf{G}^{\#}$ KJ mol ⁻¹	Ea
293	57.21	-133.30	96.26	50 65 VI
298	57.16	-136.14	97.73	$(14.256 \times 10^3 \text{ Calorian})$
303	57.12	-130.70	96.72	(14.250 x 10 Calones)

Table 5. Thermodynamic parameters for the degradation of MR

Reaction with Zirconium Graphite Modified electrode (Zr/GME) as Anode Electrode

Effect of Methyl Red on the rate: The experiments were carried out at five different concentrations varying $(0.5 \times 10^{-4} \text{ to } 2.0 \times 10^{-4} \text{ N})$ and keeping current constant (4.0 mA). The change in concentration of the MR was recorded by change in color using spectrophotometer. This study indicates as the concentration of dye is increased, the rate of the reaction is decreased and the rate constant values are higher compare to graphite electrode. A plot of log% T (percentage transmittance of light) versus time in min was linear up to 60% reaction indicating disappearance of MR follows 1st order kinetics. The pH value before and after the degradation indicates slight increase towards alkaline pH. The rate constant values are reported in table 5 and figure 6 (A).The COD values for MR solution before and after degradation were measured and are reported in table 5 and figure 6 (B).

 Table 5. Effect of concentration MR on the rate of degradation and COD values by Zr/GME electrode
 [Applied Current = 4.0mA, Temperature = 298K]

10 ⁻⁴	10^4 k	Effect of pH		COD Value	es in mg L ⁻¹
[MR] in N	in Sec ⁻¹	Before degradation	After degradation	Before degradation	After degradation
0.5	2.303	6.15	6.05	416	48
1.0	1.650	6.19	5.97	480	16
1.5	1.420	5.99	5.82	576	32
2.0	0.997	5.57	5.52	608	44



Figure 6. [A] Effect of concentration of MR dye on the rate of degradation and [B] COD values.

Effect of current on the rate: To investigate the effect of current, the experiments were carried out at four different current varying from 2.0 mA-5.0 mA and keeping [MR dye] constant. This study indicates the increase of current increases the rate of the reaction. The rate of degradation was higher

compared to graphite electrode. The rate constant values are reported in table 6 and figure 7 (A). The COD values for before and after degradation were measured and reported in table 6 and figure 7(B).

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Commont in	10 ⁴ lr	COD Value	es in mg L ⁻¹
mA	In Sec ⁻¹	Before degradation	After degradation
2.0	0.882	480	32
3.0	1.305	480	32
4.0	1.650	480	16
5.0	2.111	480	38

Table 6. Effect of current on the rate of degradation and COD values $[MR = 1.0X10^{-4} \text{ in N}, \text{ Temperature} = 298 \text{K}]$



Figure 7. [A] Effect of current of MR on the rate of degradation and [B] COD Values.

Effect of temperature: To examine the effect of temperature, the experiments were carried out at three different temperatures. It is clear that, the increase of temperature increases the rate of the reaction, and observed that, the rate of removal of degradation is not very significant at low temperature. However the reaction was more significantly influenced at higher temperature. The rate constant values and COD values for before and after degradation are reported in table 7 and figure 8(A and B). Since the diffusion rate is increased with temperature, an increase of temperature could bring about an increase in the degradation rate. Thermodynamic parameters for the degradation of MR by Zr/GME electrode were measured and reported in table 8.

Tomporatura	10 ⁴ k	COD Value	es in mg L ⁻¹
in K	In Sec ⁻¹	Before degradation	After degradation
293	1.22	480	32
298	1.65	480	16
303	2.49	480	34

Table 7. Effect of temperature on the rate of degradation and COD values $[MR = 1.0X10^{-4} \text{ in N}, \text{ Applied Current} = 4.0\text{mA}]$



Figure 8. A] Effect of temperature of MR on the rate of degradation and [B] COD Values

Table 8. 7	Fhermodynamic	parameters for	the degradation	of MR by Z	Zr/GME electrode
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Temperature	$\Delta \mathbf{H}^{\#}$	$\Delta S^{\#}$	$\Delta \mathbf{G}^{\#}$	Ea
in K	KJ mol ⁻¹	J ⁻¹ K ⁻¹ mol ⁻¹	KJ mol ⁻¹	
293	32.01	-210.41	93.66	34.45KJ
298	31.97	-210.07	94.57	(8.234 x 10 ³ Calories)
303	31.93	-208.69	95.16	

Production of OH radical: The degradation of MR solutions were carried out by anodic oxidation. Further, the experiment is carried out in the presence of electrodes with Pt as cathode and graphite as anode. In the electrochemical technique OH radical formed by direct electrolysis absorbed as intermediate at the surface of high O_2 overvoltage anode from oxidation of water.

$$H_2O \longrightarrow OH_{ads} + H^+ e^-$$

The degradation of MR takes place due to sequential electrochemical reduction and oxidation. Graphite electrodes have high affinity for MR with implication that it will adversely affect electrode performance by competitive adsorption and interaction such as electron shuttling may increase the reactivity. The strong oxidizing agent 'OH radical, the sequential reduction/oxidation with graphite and Pt electrodes which reacts with MR converting into CO_2 , NO_2 , H_2O and inorganic ions until their total mineralization were reached. It has been found that Pt electrode possesses greater O_2 over voltage (+0.77V), thus generating higher amount of oxidant 'OH.A clear solution was obtained after degradation indicates the absence of formation of insoluble metal oxides. This suggests that oxidizing intermediates which are formed resulting in destruction of the dye. Considering the graphite electrode (GE) advantages such as commercial availability, low costs and easy of modification [16, 17], the present method can be used for remediation of waste water.

Effect on COD of duration of treatment: The effect of rate of dye degradation on COD was determined. During the experiment the initial COD 416 mg L^{-1} which gave rise to 48 mg L^{-1} of COD, a 95% reduction in COD was achieved. The COD level obtained after degradation was within the permitted limit of 250 mg L^{-1} . The effect of current density, [dye] and COD was measured. In all



Figure 9. Degradation pathway of Methyl Red.

these cases the COD level decreased more than 98 %. A graph showing COD reduction versus treatment time is shown in figure 10.



Figure 10. The rate of decrease of COD at different intervals of time

Calculation of the instantaneous current efficiency (ICE): The instantaneous current efficiency (ICE) for the anodic oxidation of Methyl Red dye was calculated from the values of the COD using the equation.

ICE (%)=
$$\frac{(\text{CODi}-\text{CODf})}{8It} \times \text{FV}$$

Here; $COD_{initial}$ and COD_{final} are the chemical oxygen demands before and after degradation respectively, 'I' the current, 'F' the Faraday constant, 'V' the volume of the electrolyte, 't' the

treatment time and '8' is the oxygen equivalent mass (geq-1). The ICE data indicates the efficiency of the process linearly related to ICE and also indicates higher efficiency of the process for Zr/GME anode compared to graphite electrode anode and is reported in tables 9 and 10.

Table 9. I	CE values a	different	experimental	conditions for	Graphite	Electrode
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	Variables	ICE
Concentration of Dye 10 ⁻⁴	0.5	221.95
	1.0	109.08
	1.5	71.22
	2.0	52.33
	2.0	173.70
	3.0	123.17
Current in mA	4.0	109.08
	5.0	101.57
	293	135.10
Temperature in K	298	109.08
	303	207.84

Table	10.	ICE	values	at different	experimental	conditions	for 2	Zr/GME	E electrode
					1				

	Variables	ICE
4	0.5	443.90
Concentration of Dye 10 ⁻⁴	1.0	215.26
	1.5	154.40
	2.0	75.31
	2.0	284.42
	3.0	225.16
Current in mA	4.0	215.26
	5.0	231.60
	293	150.11
Temperature in K	298	215.26
	303	372.21

Kinetics of Degradation: The degradation of dye took place in the absence of electrolyte HCl or NaOH. The degradation of dye depends with [dye], current (*I*) and the concentration of surface active sites[S].Since[S]remains constant, the rate of degradation in the present case is given by

$$-\frac{dc}{dt} = \frac{k[I]}{[dye]}$$

The rate constant for the disappearance of Methyl Red for the degradation process was determined by plotting log T versus time (t), indicating kinetics follows 1st order reaction. Here 'T', is percentage transmission of the light obtained from spectrophotometer, at λ_{max} 410 nm and it is inversely proportional to the concentration of the dye. The straight line was obtained for 60% of the degradation reaction and afterwards deviation in linearity was observed.

Reuse of Zirconium graphite Modified electrode (Zr/GME): The possibility of reusing the Zr/graphite modified electrode was tested to see the cost effectiveness of the method adopted. After the degradation of the dye, the graphite modified electrode was thoroughly washed with double

distilled water and then reused for the degradation by taking fresh dye solution. From the degradation study it was observed that the graphite modified electrode can be used to degrade the dye solution showed, with slightly lesser efficiency.

UV-Visible spectra: Figure 11 (A and B) represents UV-Visible spectra during the electrochemical degradation of Methyl Red dye. Broad visible colour absorption spectra at 410 nm were completely removed at the end of the degradation process which indicates the decomposition of MR.



Figure 11: UV-Visible spectra of MR [A] before degradation [B] After degradation.

Scanning Electron Microscopy (SEM): Figure 12 and 13 shows the SEM image and its corresponding EDX profile has shown. From the SEM micrograph, the layered and homogenous nature of graphite was observed in different diameter. The observed size of graphite flake was ranging from 10 to 50 μ m. From the EDX profile, the presence of only carbon content indicated the graphite present in pure form. Quantitative results for graphite electrode table were reported in table 11.

Figure 14 and 15 shows the SEM and EDX profile of Zirconium encapsulated in graphite rod. The SEM image of Zirconium encapsulated graphite rod can clearly distinguish from graphite rod. Further, like heterogeneous glass pieces were observed on the surface, which depict the Zirconium doped or encapsulated inside the graphite rod. The corresponding EDX plot showed in figure 15. From the EDX plot, we clearly noticed the presence of Zirconium peaks in different oxidation states along with graphite carbon peak. Quantitative results for Zirconium graphite modified electrode are reported in table 12.



Figure 12. SEM micrographs of graphite electrode.



Figure 13(B). Energy –dispersive X-ray spectroscopy (EDX) for graphite Electrode.

Table 11. Quantitative results for graphite Electrode

Element Line	Weight %	Weight % Error	Atom %
C K	100.00	± 1.09	100.00
Total	100.00		100.00



Figure 14. SEM micrographs of Zr/ graphite modified electrode.



Figure 15. Energy –dispersive X-ray spectroscopy (EDX) for Zr/graphite modified electrode.

Element Line	Weight %	Weight % Error	Atom %
C K			
O K	10.29	± 0.72	42.00
Zr L	89.71	± 1.51	58.00
Zr M			
Total	100.00		100.00

Table 12. Quantitative results for Zr/graphite modified electrode

APPLICATION

This can be used for wastewater treatment and purification of industrial effluents.

CONCLUSION

In this paper, electrochemical oxidation in presence of graphite electrode and Zr/GME was capable of destroying the chromophore groups of dye found in industrial effluents at short treatment times, low energy consumption. This method can be applied to the remediation of wastewater containing dyes and organics and the method is cost effective as graphite electrode was used. The rates of MR elimination and COD removal were faster on the Zr/GME compare to graphite electrode and it can be reused.

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