



Contents lists available at ScienceDirect

Process Safety and Environmental Protection

journal homepage: [www.elsevier.com/locate/psep](http://www.elsevier.com/locate/psep)

IChemE

## Destruction of methylene blue by mediated electrolysis using two-phase system

Manickam Matheswaran<sup>a,\*</sup>, T. Raju<sup>b</sup>

<sup>a</sup> Department of Chemical Engineering, National Institute of Technology, Tiruchirappalli 620 015, Tamilnadu, India

<sup>b</sup> Central Electrochemical Research Institute, Karaikudi 630006, India

### A B S T R A C T

The destruction and colour removal of methylene blue were carried out by mediated electrochemical oxidation using cerium(IV) mediator in nitric acid medium using two-phase system and was applied for oxidation of organic compound. Organic compounds and mediator were taken in the organic and aqueous phases respectively. The influence of organic solvent type, cerium(III) concentration, dye concentration, stirring speed and temperature were investigated in order to find the optimum conditions of the system to check the removal of COD and colour in the organic phase. The decolourisation followed the pseudo-first order reaction for electrochemically oxidized cerium(IV). The maximum colour removal efficiency of 95% was achieved within 45 min in dichloromethane solvent system. The COD removal efficiency and colour removal rate were increased with increase in temperature and stirring speed.

© 2010 The Institution of Chemical Engineers. Published by Elsevier B.V. All rights reserved.

**Keywords:** Electrochemical oxidation; Methylene blue; Two-phase system; Cerium(IV); Mediator

### 1. Introduction

Environmental pollution especially by the dying and textile industries is one of the major problems faced by the developing countries like India. It was reported approximately 5 tonnes of organic compounds discharge from dyes and colouration industries every year (Brown, 1987). Due to stringent environmental regulation, these wastewater has to be treated before disposal into the environment. A conventional treatment which includes biological, physical, chemical processes was employed to treat the wastewater. Removal of organic compounds from wastewater by many conventional techniques like adsorption (Matheswaran and Karunanithi, 2007; Preethi et al., 2006), coagulation (Bache and Hossain, 1991), membrane process, filtration, precipitation (Sarasa et al., 1998), advanced oxidation process (Aleboyeh et al., 2005; Mohey El-Dein et al., 2003), chemical oxidation, etc. showed better performance, but the production of secondary waste like sludge, increases treatment cost. Hence, the treatment of wastewater economically is becoming a matter of great concern and it is urgent to develop the cost-effective treatment technologies to reach related standards before being discharged into the environment.

Electrochemical techniques, a powerful means of pollution control, are considered as an attractive methodology for the treatment of dyeing wastewater and have significant advantages namely: wide application, simple equipment, easy operation, lower temperature requirements and no sludge formation (Daneshvar et al., 2004; Gutierrez and Crespi, 1999). Several researchers have investigated the feasibility of electrochemical degradation of dye with various electrode materials for wastewater treatment, such as titanium-based dimensionally stable anode (DSA) (Szpyrkowicz et al., 2000), platinum (Sanroman et al., 2004), conductive-diamond (Canizares et al., 2006), metal alloy (Rivera et al., 2004) and boron-doped diamond (BDD) (Fernandes et al., 2004). During high potential electrolysis, these electrodes produce hydroxyl radicals from the water discharge on their surfaces. With regard to these electrodes, common drawbacks of SnO<sub>2</sub> and PbO<sub>2</sub> are a short service-life (Correa-Lozano et al., 1997) and the release of toxic ions, while BDD anodes have good chemical and electrochemical stabilities even in strong aggressive media, long life, and a wide potential window for water discharge. Thus, these electrodes were not promising anodes for industrial-scale wastewater treatment and the cost of the electrodes and chemicals employed are high (Panizza and Cerisola, 2008).

\* Corresponding author. Tel.: +91 431 2503101.

E-mail address: [math.chem95@rediffmail.com](mailto:math.chem95@rediffmail.com) (M. Matheswaran).

Received 20 July 2009; Received in revised form 17 May 2010; Accepted 25 May 2010

0957-5820/\$ – see front matter © 2010 The Institution of Chemical Engineers. Published by Elsevier B.V. All rights reserved.

doi:10.1016/j.psep.2010.05.003

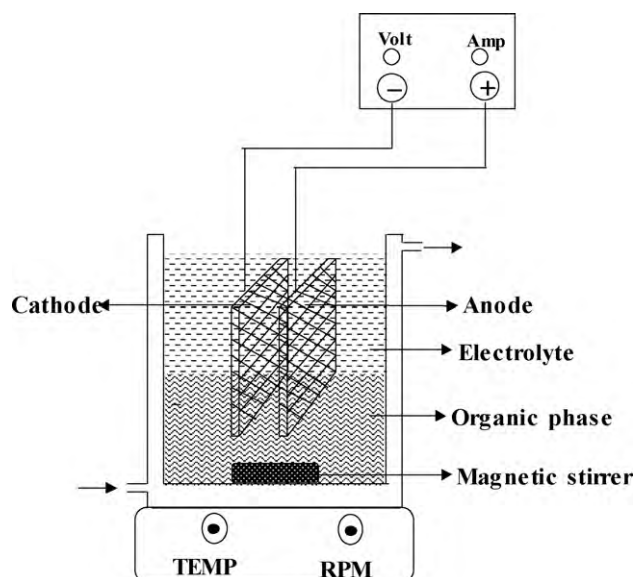


Fig. 1 – Schematic diagram of experimental setup.

Therefore, it is necessary to find an effective method of wastewater treatment that are capable of removing colour and degrading toxic organic compounds from industrial effluents. The electric current induces redox reactions upon the electrodes surface resulting in the destruction of the organic compound. It makes the treatment of liquids, gases and solids possible; it is compatible with the environment because the electron is one of the main reagents, i.e. clean and safe (Juttner et al., 2008). Hence, electron is an environmental friendly and cost competitive alternative.

Mediated electrochemical oxidation (MEO) is one such promising technology for destruction of hazardous organic waste and also applied for dissolution of metal in the military waste. In this process, oxidized state of mediator destructs the organic compounds into carbon dioxide and water, then it reduced. The reduced state redox mediator is continuously regenerated by electrochemical cell (Farmer et al., 1992; Matheswaran et al., 2007). Different types of mediators silver, cerium, cobalt, manganese, etc. were used for various application like destruction of organic, degradation of aromatic liquids and mineralization of effluents, and electro synthesis of organic compounds. In our study, Ce(III)/Ce(IV) redox system is used as a mediator in the medium which has high solubility and potential of 1.72 V in the nitric acid medium. Katafias et al. (2007) studied oxidation of methylene blue (MB) by Ce(IV) ions in sulphuric acid medium. They reported on kinetic data for irreversible oxidation. Donaldson et al. (2002) reported on anodic oxidation of various dyes using NaCl as an electrolyte medium. During the destruction, various intermediate and chlorinated compounds formed may be more harmful to aqueous environment than the dye molecule from wastewater. Hence, the dye molecule needs to be destructed into carbon dioxide and water. The cobalt redox mediator was used for the decolourisation of dyes of various structures and reviewed by Sanroman et al. (2004).

By using the redox mediator, the dye can be destructed completely into carbon dioxide and water at room temperature conditions. The MB was oxidized and decolourized with Ce(III)/Ce(IV) redox mediator in two-phase system. The major advantage of two-phase system was maintaining the redox mediator concentration without any dilution during the electrochemical oxidation and regenerated mediators con-

tinuously. The organic solvent can be reused for further experiments with negligible loss. During the destruction of organic compound, any useful intermediate can be easily recovered by means of extraction.

The objective of the present work is to study the decolourisation and destruction of MB by Ce(IV) mediated electrochemical oxidation by two-phase system. Various solvent mediums were investigated for decolourisation. Also the effect of various operating parameters such as concentration of Ce(III), concentration of MB, stirring speed and temperature on COD and colour removal percentage were studied. The rate of colour removal was analyzed by applying the pseudo-first order rate equation. The effect of various organic solvents on the extraction of MB from the aqueous solution was studied at a known concentration.

## 2. Experimental

### 2.1. Materials

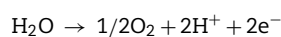
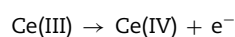
Methylene blue (purity >98%) was purchased from the Duksan Pharmaceutical Co. Ltd., and used without further purification. A solution of MB was prepared and dissolved in organic solvent. Cerium(III)nitrate hexahydrate (Terio Corporation, China), nitric acid (60%) (Sam Chun Chemicals, Korea), dichloromethane (DCM), dichloroethane (DCE), chloroform, acetonitrile (Kanto Chemical Co. Inc.) were used as received.

### 2.2. Experimental method

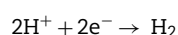
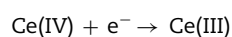
The electrochemical oxidation of MB was carried out in a batch type undivided electrochemical cell. DSA and titanium mesh type electrodes were used as the anode and cathode respectively with area of 4 cm<sup>2</sup> (2 cm × 2 cm) and both the electrodes were placed vertically and parallel to each other in the electrolytic cell. The anode and cathode have an inter-electrode gap of 2 cm. The schematic diagram of electrochemical cell setup is shown in Fig. 1. 0.4 A of current was passed during the electrolysis experiments which was provided by a laboratory DC power supply (KSC, Korea Switching). A 100 ml volume cell constituting 50 ml of electrolyte {Ce(III) and 3 M nitric acid} and 50 ml of organic solvent that contains MB with known concentration were used. The electrolytic cell was equipped with a magnetic stirrer in order to keep the electrolyte and organic solvent well mixed for effective reaction. The temperature of the system was maintained constant during the electrolysis. During the oxidation, Ce(III) was oxidized into Ce(IV) at an anode. The oxidized Ce(IV) reacted with MB in the organic solvent, then Ce(IV) reduced into Ce(III) and continuously regenerated by electrochemical cell.

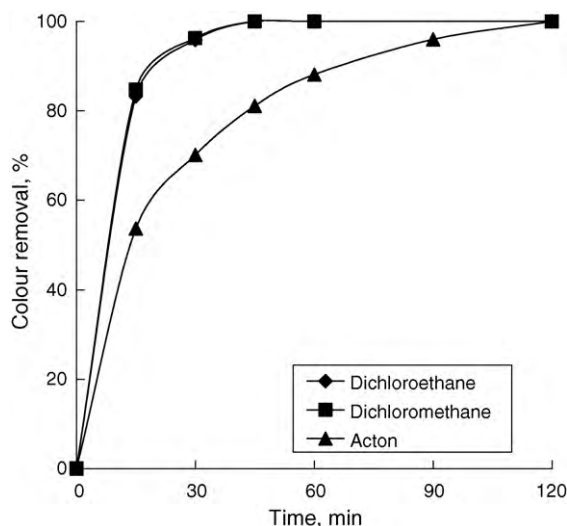
The following reaction can take place in the cell while passing constant current (Sedneva, 2005):

Anode:



Cathode:





**Fig. 2 – Effect of organic solvent on colour removal of MB** (conditions:  $C_{\text{Ce(III)}}$ : 0.25 M;  $C_{\text{HNO}_3}$ : 3 M; current: 0.4 A;  $C_{\text{MB}}$ : 0.25 M; stirring rate: 500 rpm).

The samples of organic solvent were taken at different intervals of time over a period of 2 h for analyzing colour removal and chemical oxygen demand (COD). The UV–vis spectrophotometer (UV, model 160A, Shimadzu) was used to measure the concentration of MB at 662 nm.  $\text{COD}_{\text{Cr}}$  was measured with a COD reactor and spectrometer by using HACH procedure (Balaji et al., 2007a,b). The effect of organic solvent, concentration of Ce(III), agitation speed and temperature were investigated based on the removal of colour and COD in the two-phase system. The effect of solvent on the extraction efficiency of MB from the aqueous solution was carried out. A known concentration of 0.5 mM MB was taken in the 50 ml of aqueous solution and mixed with 50 ml of organic solvent. The mixture was thoroughly mixed. The organic and aqueous phases were taken for analysis of MB after it reaches the phase separation.

From the absorbance values, the colour removal efficiency was calculated using the following formulae:

$$\text{Colour removal efficiency} = \frac{\text{ABS}_{\text{Mo}} - \text{ABS}_{\text{M}}}{\text{ABS}_{\text{Mo}}} \times 100$$

where  $\text{ABS}_{\text{M}}$  is the average of absorbance values, at its maximum absorbance value of visible wavelength ( $\lambda_{\text{max}}$ ).  $\text{ABS}_{\text{Mo}}$  and  $\text{ABS}_{\text{M}}$  are the values before and after the degradation process.

The COD removal efficiency ( $\eta$ ) was calculated using the following formula:

$$\eta = \frac{C_0 - C_t}{C_0} \times 100$$

where  $C_0$  is the COD of initial MB concentration, and  $C_t$  is the COD of the remaining dye concentration at given time  $t$ .

### 3. Results and discussions

#### 3.1. Effect of organic solvent

Studies were carried out for the effect and selection of suitable solvent system for the decolourisation of MB by electrochemically generated Ce(IV) redox mediators. The results are given in Fig. 2. Various types of organic solvents such as DCE, DCM, acetonitrile and chloroform were studied. Mostly, in

two-phase electrolysis studies chloroform has been used as solvent (Raju et al., 2006). In these studies, where chloroform was used as solvent with electrochemically generated Ce(IV) redox mediators it is found that the MB dyes phase transfer from organic solvent to electrolyte while mixing. Selection of solvent also depends on the type of electrolyte used for the oxidation of organic compound. When the DCE and DCM were used as solvent, 95% of removal efficiency of MB was achieved. Acetonitrile solvent also gave good removal efficiency of 95% for decolourisation of MB. But it resulted in an emulsion, which is not preferred.

Effect of solvents on the extent of solubility, rates of mass transfer and decolourisation of MB may be related to their thermodynamic and physical properties such as dielectric constants, dipole moments, polarity index density and viscosity. This effect may be attributed to improvement of mass transfer rate due to enhanced solubility of MB by the solvents. While the aqueous phase concentrations of residual Ce(IV) were more for all the solvents, the concentrations of sorbed MB were substantially higher for solvents employed in this system.

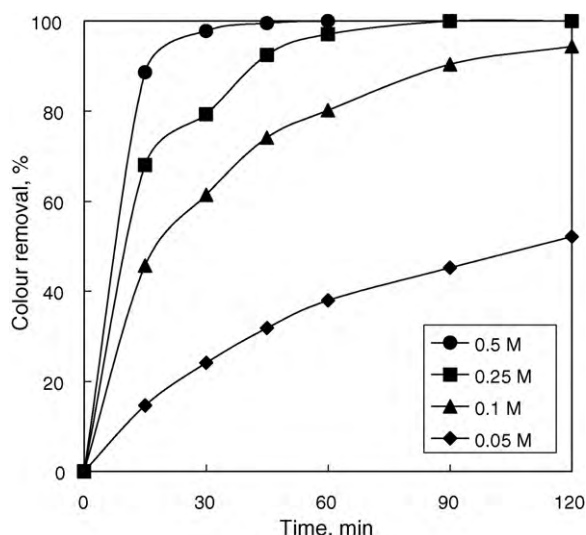
In the present investigation, efficiencies of removal of MB by MEO system correlated well with the dipole moments of solvents. It is evident that the maximum decolourisation of MB was achieved by Ce(III)/Ce(IV) system using DCM or DCE as a solvent, which had the highest dielectric constants and dipole moment. It may be noted that dipole moments for the solvents were very close to each other (1.15–1.8) and dielectric constants of the DCM (8.93) and DCE (10.36) are high and so were the concentrations of residual MB after 45 min of reaction, it completely decolourised. Morita et al. (1988) studied chlorination of toluene by two-phase electrolysis using different solvents for production of o- and p- chlorotoluene and observed that the percentage yield was increased in DCM and DCE compared to other solvents.

Extraction of 0.5 mM MB from 50 ml of aqueous solution using various solvents of DCM, DCE and chloroform was carried out for studying the removal efficiency. The concentration (removal efficiency) of MB in the aqueous phase was 0.035 mM (93%), 0.028 mM (94%) and 0.26 mM (50%) using solvents of DCM, DCE and chloroform respectively.

#### 3.2. Effect of cerium(III) concentration

The influence of cerium concentration on the removal of colour and destruction of MB were studied. Fig. 3 shows the colour removal efficiency of MB as a function of time at different initial concentrations of Ce(III) in the aqueous phase of electrolyte at fixed concentration of nitric acid (3M), temperature (25 °C) and current (0.4 A). It was observed from the figure, colour removal was increased with increase in concentration of Ce(III). When the oxidation rate of Ce(III) increased with increase in the initial concentration of Ce(III), the formation of Ce(IV) was found to increase. The destruction of MB was measured in terms of COD removal after 2 h. The percentage of COD removal increases from 17 to 60% while increasing the concentration of Ce(III) from 0.05 M to 0.5 M in 3M nitric acid. The percentage removal of colour was 100% within 90 min in case of 0.25 M and also for higher concentrations of Ce(III). However the removal of COD increased when the concentration of Ce(III) increased from 0.25 M to 0.5 M.

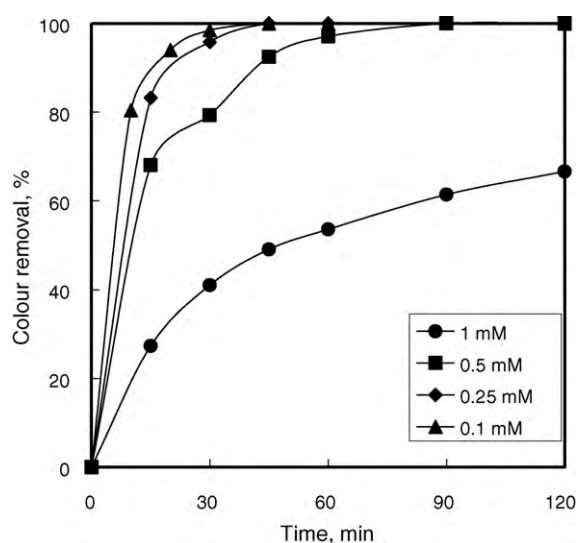




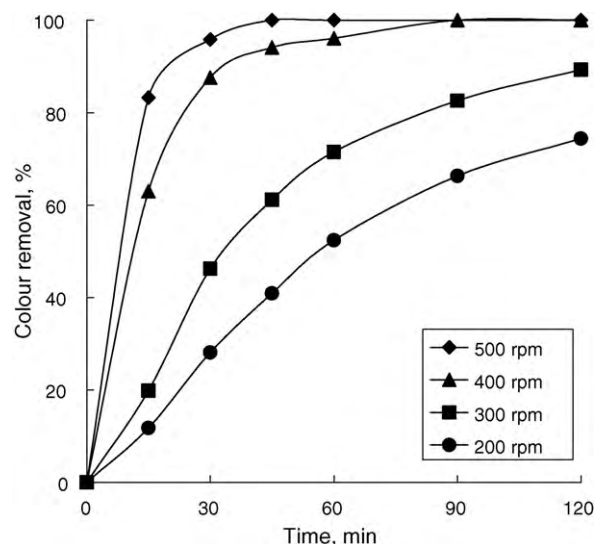
**Fig. 3 – Effect of cerium(III) concentration on colour removal of MB (conditions:  $C_{HNO_3}$  : 3 M; T: 20 °C; current: 0.4 A;  $C_{MB}$ : 0.5 mM; stirring speed: 500 rpm).**

### 3.3. Effect of initial MB concentration

The initial dye concentration is one of the crucial parameter in any electrochemical treatment. The effect of initial concentration of MB in the organic solvent was studied using electrochemical oxidation by varying the MB concentrations from 0.1 mM to 1 mM under fixed concentration of Ce(III) (0.25 M) and current (0.4 A). Fig. 4 shows the percentage of colour removal as a function of time. The rate of colour removal was decreased with increase in initial concentration of MB with a maximum of 100% upto 0.25 mM within 1 h. Similar results observed by Zhou et al. (2008) using electrochemical oxidation of various dyes like C.I. Acid Red 2, methyl red. The percentage of COD removal in the organic solvent was decreased as the initial concentration increases. Mohey El-Dein et al. (2003) reported that decolourisation rate of azo dye by UV and  $H_2O_2$  followed the first order with respect to dye concentration.



**Fig. 4 – Effect of initial MB concentration on colour removal of MB (conditions:  $C_{Ce(III)}$ : 0.25 M;  $C_{HNO_3}$  : 3 M; T: 20 °C; current: 0.4 A; stirring speed: 500 rpm).**



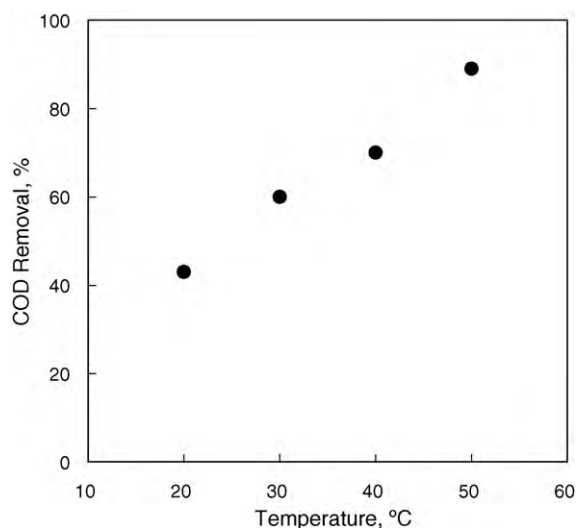
**Fig. 5 – Effect of stirring speed on colour removal of MB (conditions:  $C_{Ce(III)}$ : 0.25 M;  $C_{HNO_3}$  : 3 M; T: 20 °C; current: 0.4 A).**

### 3.4. Effect of agitation rate

Fig. 5 shows the colour removal percentage of different stirring rates at fixed concentration of Ce(III) and dye, nitric acid and electrolysis temperature. It was observed that removal of colour was increased with increase in stirring speed. A maximum removal of 100% was achieved at 500 rpm in 1 h. It might be due to the mass transfer rate enhancement at a speed of 500 rpm than the other. Hence, the destruction of dye is faster at high agitation rate when compared to the low agitation rate and also the reaction between cerium and MB was carried out efficiently by increasing the interfacial area of aqueous and organic phases. The pseudo-first order rate constant for colour removal was varied from  $0.0117 \text{ min}^{-1}$  to  $0.1084 \text{ min}^{-1}$ . The percentage of COD removal was increased from 11 to 43% with increasing stirring rate from 200 rpm to 500 rpm. From the studies, it is found that the cell voltage was low at slow stirring rate of the electrolyte solution, when the stirring rate is increased, cell voltage also increased considerably. It might be due to the phase state of the system. At lower stirring rate the aqueous and organic solutions are in the two separated phase but while increasing the stirring rate the solution is in the emulsion form. So the resistance increased due to the electrode contact of organic phase.

### 3.5. Effect of temperature

To study the influence of temperature on the electrochemical oxidation process, temperature of the solution was varied from 20 °C to 50 °C at 0.25 M Ce(III) and 0.25 mM of MB. The percentage of COD removal was 43–85% while increasing the temperature from 20 °C to 50 °C as shown in Fig. 6. Temperature exhibited an acceleration of electrochemical generation of Ce(IV). Increase in the temperature, increases the removal percentage of colour and COD for MB in the organic solvent. Similar trend was also observed by Balaji et al. (2007a,b) in the Ce(IV)-MEO. They reported that the destruction efficiency of phenol was increased with increasing temperature. The colour removal efficiency was observed nearly 100% with 45 min for all temperatures. Specific energy consumption decreased with



**Fig. 6 – Effect of temperature on COD removal of MB**  
(conditions:  $C_{\text{Ce(III)}}$ : 0.25 M;  $C_{\text{HNO}_3}$ : 3 M; current: 0.4 A;  $C_{\text{MB}}$ : 0.25 M; stirring rate: 500 rpm).

increase in temperature due to cell voltage decrease during the electrochemical oxidation.

#### 4. Conclusion

Decolourisation and destruction of MB were studied with the Ce(IV) redox mediators by two-phase mediated electrochemical oxidation. DCM and DCE were suitable solvent for the system. The extraction efficiency of MB from aqueous solution using solvents DCM and DCE were 93 and 95% respectively. The Ce(IV) was used as mediator in nitric acid medium successfully in oxidation of MB. The effect of various operating parameters was studied for the removal of MB colour. The percentage of COD removal and rate of colour removal of MB were increased with increasing concentration of Ce(III) and electrolyte temperature. The colour removal efficiency was observed 100% within 45 min duration. The stirring rate increases the interface between Ce(III) and MB, which increases the colour and COD removal percentage.

The present technique showed a stable colour and COD removal in a wider dye concentration ranges, representing the most promising alternative for environmental application among the conventional processes. There is no consumption of chemicals in two-phase electrolysis system as compared to the conventional chemical methods for decolourisation process. The redox mediator Ce(III)/Ce(IV) can be regenerated and reused.

#### References

Aleboye, A., Moussa, Y. and Aleboye, H., 2005, Kinetics of oxidative decolourisation of Acid Orange 7 in water by ultraviolet radiation in the presence of hydrogen peroxide. *Sep. Purif. Technol.*, 43: 143–148.

Bache, D.H. and Hossain, M.D., 1991, Optimum coagulation conditions for coloured water in terms of floc properties. *J. Water Services Res. Technol.—Aqua*, 40: 170–178.

Balaji, S., Chung, S.J., Matheswaran, M. and Moon, I.S., 2007a, Cerium(IV) mediated electrochemical oxidation process for the destruction of various organic pollutants in batch and continuous organic feeding modes. *Korean J. Chem. Eng.*, 24: 1009–1016.

Balaji, S., Chung, S.J., Matheswaran, M. and Moon, I.S., 2007b, Cerium(IV)-mediated electrochemical oxidation process for destruction of organic pollutants in a batch and a continuous flow reactor. *Korean J. Chem. Eng.*, 24: 1009–1016.

Brown, D., 1987, Effects of colorants in the aquatic environment. *Ecotoxicol. Environ.*, 13: 139–147.

Canizares, P., Gadri, A., Lobato, J., Nasr, B., Paz, R., Rodrigo, M.A. and Saez, C., 2006, Electrochemical oxidation of Azoic Dyes with conductive-diamond anodes. *Ind. Eng. Chem. Res.*, 45: 3468–3473.

Correa-Lozano, B., Comninellis, C. and DeBattisti, A., 1997, Service life of Ti/SnO<sub>2</sub>-Sb<sub>2</sub>O<sub>5</sub> anodes. *J. Appl. Electrochem.*, 27: 970–974.

Daneshvar, N., Sorkhabi, H.A. and Kobya, M., 2004, Decolorization of reactive dye solution containing Acid Red 14 by electrocoagulation with a comparative investigation of different electrode connections. *J. Hazard. Mater.*, 112: 55–62.

Donaldson, J.D., Grimes, S.M., Yasri, N.G., Wheals, B., Parrick, J. and Errington, W.E., 2002, Anodic oxidation of the dye materials methylene blue, acid blue 25, reactive blue 2 and reactive blue 15 and the characterization of novel intermediate compounds in the anodic oxidation of methylene blue. *J. Chem. Technol. Biotechnol.*, 77: 756–760.

Farmer, J.C., Wang, F.T., Hawley-Fedder, R.A., Lewis, P.R., Summers, L.J. and Foiles, L., 1992, Electrochemical treatment of mixed and hazardous wastes: oxidation of ethylene glycol and benzene by silver(II). *J. Electrochem. Soc.*, 139: 654–661.

Fernandes, A., Morao, A., Magrinho, M. and Goncalves, I., 2004, Electrochemical degradation of C.I. Acid Orange 7. *Dyes Pigm.*, 61: 287–296.

Gutierrez, M.C. and Crespi, M., 1999, A review of electrochemical treatments for color elimination. *J. Soc. Dyes Colour.*, 115: 342–345.

Juttner, K., Galla, U. and Schmieder, H., 2008, Electrochemical approaches to environmental problems in the process industry. *Electrochim. Acta.*, 45: 2575–2594.

Katafias, A., Kita, P., Wrzeszcz, G. and Mills, A., 2007, Kinetics of the methylene blue oxidation by cerium(IV) in sulphuric acid solutions. *Transition Met. Chem.*, 32: 31–37.

Matheswaran, M., Balaji, S., Chung, S.J. and Moon, I.S., 2007, Silver mediated electrochemical oxidation: production of silver(II) in nitric acid medium and destruction of phenol in semi batch process. *J. Ind. Eng. Chem.*, 13: 231–236.

Matheswaran, M. and Karunanithi, T., 2007, Adsorption of Chrysoidine R by using fly ash in batch process. *J. Hazard. Mater.*, 145: 154–161.

Mohey El-Dein, A., Libra, J.A. and Wiesmann, U., 2003, Mechanism and kinetic model for the decolorization of the azo dye Reactive Black 5 by hydrogen peroxide and UV radiation. *Chemosphere*, 52: 1069–1077.

Morita, M., Yamamoto, S. and Matsuda, Y., 1988, Chlorination of toluene by two-phase electrolysis. *J. Appl. Electrochem.*, 18: 491–492.

Panizza, M. and Cerisola, G., 2008, Removal of colour and COD from wastewater containing acid blue 22 by electrochemical oxidation. *J. Hazard. Mater.*, 153: 83–88.

Preethi, S., Sivasamy, A., Sivanesan, S., Ramamurthi, V. and Swaminathan, G., 2006, Removal of Safranin Basic Dye from aqueous solutions by adsorption onto Corn cob activated carbon. *Ind. Eng. Chem. Res.*, 45: 7627–7632.

Raju, T., Kalpana Devi, G. and Kulangiappar, K., 2006, Regioselective bromination of toluene by electrochemical method. *Electrochim. Acta.*, 51: 4596–4600.

Rivera, M.C., Jimenez, M.M.D. and Gonzalez, M.P.E., 2004, Degradation of the textile dyes Basic yellow 28 and Reactive black 5 using diamond and metal alloys electrodes. *Chemosphere*, 55: 1–10.

Sanroman, M.A., Pazos, M., Ricart, M.T. and Cameselle, C., 2004, Electrochemical decolourisation of structurally different dyes. *Chemosphere*, 57: 233–239.

- Sarasa, J., Roche, M.P., Ormad, M.P., Gimeno, E., Puig, A. and Ovelheiro, J.L., 1998, Treatment of a wastewater resulting from dyes manufacturing with ozone and chemical coagulation. *Water Res.*, 32: 2721–2727.
- Sedneva, T.A., 2005, Cerium(III) Electrooxidation in Nitric Acid Solutions of Rare-earth Elements. *Russ. J. Appl. Chem.*, 76: 907–911.
- Szpyrkowicz, L., Juzzolino, C., Kaul, S.N., Daniele, S. and Faveri, M.D.D., 2000, Electrochemical oxidation of dyeing baths bearing disperse dyes. *Ind. Eng. Chem. Res.*, 39: 3241–3248.
- Zhou, M., Yu, Q. and Lei, L., 2008, The preparation and characterization of a graphite–PTFE cathode system for the decolorization of C.I. Acid Red 2. *Dyes Pigm.*, 77: 129–136.