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Electrocoagulation of Azo-2-Naphthol Dye Using Aluminium Electrodes: Effect of Solvent and Kinetics of Adsorption

V.N. Mkpennie and O.U. Abakedi

Department of Chemistry, University of Uyo, P.M.B. 1017, Uyo, Nigeria

Corresponding Author: V.N. Mkpennie, Department of Chemistry, University of Uyo, P.M.B. 1017, Uyo, Nigeria

ABSTRACT

1-Phenylazo-2-naphthol (PAN) was subjected to electrocoagulation using a 0.3 A d.c electrolytic cell in various solvents including ethanol, acetone, benzene and n-hexane. The extent of decolourization of PAN in the solvents has been determined. The highest decolourization was achieved in ethanol with colour removal efficiency of 63% within the first 15 min and 79% after 120 min of electrocoagulation. The decolourization of PAN in the solvents is observed to follow the order: Ethanol>n-hexane>benzene>acetone. The electrocoagulation data has also been interpreted based on adsorption kinetic models. Electrocoagulation of PAN in all the solvents obeyed the pseudo-second order kinetic model and Elovich kinetic model.

Key words: Electrocoagulation, aluminium electrode, azo-2-naphthol, solvent effect, adsorption kinetics

INTRODUCTION

Electrocoagulation (EC) is an electrochemical technique in which a variety of unwanted dissolved particles and suspended matters can be effectively removed from solution by electrolysis. Electrocoagulation is receiving attention because of the versatility of the technique. It is capable of fractionating a number of organic substances in efficient manner by electrochemically coagulating some of them while leaving other components in solution (Chairungsi *et al.*, 2006a). Although it was originally used as a purification step in the isolation of a few natural products from crude plant extracts (Miwa, 1979), the technique has now been successfully applied in clarifying food wastewater (Beck *et al.*, 1974; Butler *et al.*, 2011), industrial wastes containing cyanide and heavy metals (Cenkin and Belevtsev, 1985; Mrozowski and Zielinski, 1983; Jenke and Diebold, 1984), industrial wastewater containing organic pollutants (Barrera-Diaz *et al.*, 2014), tar-sand and oil-shale wastewater (Renk, 1988), portable water (Mameri *et al.*, 1998; Vik *et al.*, 1984), denitrification (Yehya *et al.*, 2014), removal of Cr(VI) from polluted solutions (Aber *et al.*, 2009), arsenic removal (Balasubramanian *et al.*, 2009) and boron removal (Isa *et al.*, 2014).

Electrocoagulation is very effective in decolourizing dye solution. It has been successfully utilized in the removal of colour from tea effluent (Maghanga *et al.*, 2009), removal of colour of spent wash (Bhise *et al.*, 2012), decolorization of methyl orange wastewater (Pi *et al.*, 2014), purification of wastewater from gelatin production plant (Kruthika *et al.*, 2013), removal of direct red 81 dye from aqueous solution (Zodi *et al.*, 2013) and treatment of dye-containing textile wastewater (Lee *et al.*, 1999; Do and Chen, 1994; Khandegar and Saroha, 2013).

The main processes involved during electrocoagulation include electrolytic reactions at the surface of electrodes, formation of coagulant in the aqueous phase and adsorption of soluble or colloidal pollutant onto coagulant and subsequent removal by sedimentation or flotation of flocs when hydrogen (H_2) bubbles are produced at the cathode (Aleboyeh *et al.*, 2008). Since adsorption is one of the major steps in electrocoagulation mechanism, adsorption models have been utilized in elucidating electrocoagulation kinetic data based on Langmuir, Freundlich and Temkin adsorption models (Ghanim and Ajjam, 2013). A Variable Order Kinetics (VOK) approach has been used to fit experimental data based on Langmuir isotherm (Hu *et al.*, 2007) and Langmuir-Freundlich model (Bennajah *et al.*, 2010). Electrocoagulation kinetics have also been tested using both pseudo first-order and pseudo second-order Lagergren rate equations (Mansour *et al.*, 2013).

Different parameters have been investigated and optimized in an attempt to fully interpret electrocoagulation processes. These include, electrode material (Bagga *et al.*, 2008; Gomes *et al.*, 2007; Chou *et al.*, 2009) inter-electrode spacing (Dalvand *et al.*, 2011), current density (Arslan-Alaton *et al.*, 2008), initial concentration/dose (Mouedhen *et al.*, 2008; Mkpenie *et al.*, 2014), initial pH (Sasson *et al.*, 2009; Irdemez *et al.*, 2006), supporting electrolyte (Trompette and Vergnes, 2009), electrolysis/treatment time (Giwa *et al.*, 2012; Avsar *et al.*, 2012) and temperature (Katal and Pahlavanzadeh, 2011; Vasudevan *et al.*, 2009a; Yilmaz *et al.*, 2008a).

A good number of electrocoagulation studies is based on aqueous medium. Electrocoagulation studies using different solvent systems are less reported. Since solvent is the medium for electrocoagulation, studying electrocoagulation processes using different solvents systems may reveal important information considering the mechanism involved. Diffusion (migration) of the *in situ* generated reactants (Al^{3+} and OH^- ions) through the solvent to form the coagulant may be enhanced by the type of solvent used since these solvents possess different properties such as refractive index, dielectric constant, polarizability, relative permittivity and dipole moment and these may affect electrocoagulation process by the solute-solvent interactions. Information on such solute-solvent interaction and how it affects electrocoagulation process is lacking.

An azo dye has been chosen for this study since it constitute one of the largest classes of industrially synthesized organic compounds that are extensively used as dye (Eaton, 1989; Mohamed and El-Din, 1999).

In this study, is reported the electrocoagulation of azo dye of 2-naphthol (1-phenylazo-2-naphthol) in different solvents, including ethanol, acetone, benzene and n-hexane. The effect of solvent on electrocoagulation is evaluated and interpretation of electrocoagulation data is given based on pseudo second-order and Elovich adsorption kinetic models.

MATERIALS AND METHODS

General: All chemicals used were of analytical grade. The 1-phenylazo-2-naphthol (PAN) was obtained from previous study (Mkpenie and Essien, 2015). Aluminium plates were purchased locally. Direct current was sustained by a GW Instek DC power supply. Absorbance was measured from a UV-Visible spectrophotometer (UV-780c).

Preparation of sample solutions for electrocoagulation: The PAN was dissolved in appropriate solvent to obtain the concentration 1.0% (w/v). The solutions were scanned through 200-800 nm wavelength using UV-Visible spectrophotometer to obtain the wavelength of maximum absorption in the various solvents. The absorption maximum, λ_{max} for the various solutions were as indicated: Polar protic solvent, ethanol ($\lambda_{max} = 400$ nm), polar aprotic solvent, acetone ($\lambda_{max} = 500$ nm), non-polar solvents, benzene ($\lambda_{max} = 600$ nm) and hexane, ($\lambda_{max} = 550$ nm).

Electrocoagulation procedures: A pair of aluminum plates (12×4 cm) spaced 3 cm apart was immersed 5.5 cm into each sample solution (250 mL) contained in a 400 mL beaker. The solution was agitated throughout the experiment with a magnetic plate stirrer with Teflon bar rotating at a velocity of 500 rpm. Sodium chloride (0.5 g) was added as supporting electrolyte. Direct current (0.3 A, 24 V) was passed through the solution via the two electrodes. The colour of the dye solution before and during the electrocoagulation was measured with a UV-Visible spectrophotometer. At 15 min interval over a 2 h period of electrolysis, 4 mL aliquot of the solution was withdrawn, centrifuged and the absorbance of the supernatant solution was measured at an appropriate wavelength corresponding to the absorption maximum for each solvent used. The data obtained were used to construct electrocoagulation curves.

RESULTS AND DISCUSSION

Electrocoagulation curves for PAN (1) in various solvents, ethanol, n-hexane, benzene and acetone were plotted as a function of colour removal efficiency versus time (Fig. 1).

Colour removal (%) was determined using the absorbance data according to Eq. 1:

$$\text{Colour removal (\%)} = 1 - A_t / A_0 \times 100 \quad (1)$$

where, A_0 and A_t are absorbance values at time zero and t (taken every 15 min), respectively. The colour removal efficiency achieved were 79% for ethanol, 66% for n-hexane, 44% for benzene and 40% for acetone (Fig. 2).

Decolourization efficiency in the solvents: Rapid electrocoagulation was observed as shown in the Fig. 2 within the first 15 min of electrolysis in all the solvents used. Rapid electrocoagulation was reported for compounds with two adjacent phenolic hydroxyl groups which seems to be particularly prone to coagulation by a metal ion, probably by complexation (Chairungsi *et al.*, 2006b). The position of the azo group and phenolic hydroxide group in azo-2-naphthol is consistent with such observation and azo group is known to possess excellent donor properties which enable them to be used in coordination chemistry (Garnovskii *et al.*, 2004). Ahlawat *et al.* (2008) also recorded rapid electrocoagulation of cotton blue acid dye within 15 min using aluminium electrodes. Ethanol showed the highest colour removal efficiency of 79% whereas, acetone recorded the least

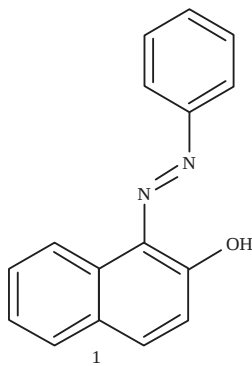


Fig. 1: Structure of 1-phenylazo-2-naphthol

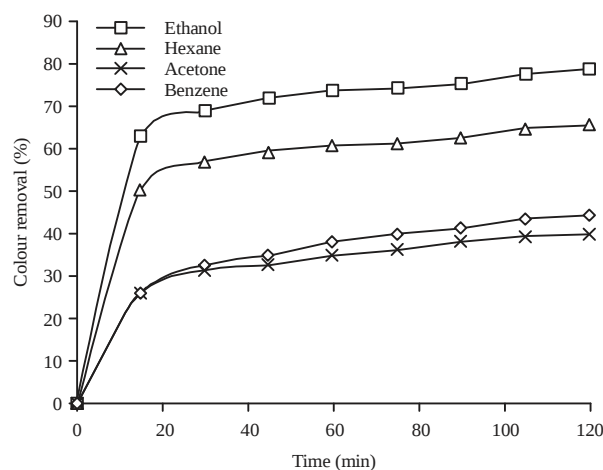


Fig. 2: Plots of colour removal efficiency vs. electrolysis time for 1.0% (w/v) of PAN in various solvents

colour removal efficiency of 40% after 120 min of electrocoagulation. Colour removal efficiency greater than 90% was obtained on electrocoagulation of textile wastewater containing disperse red dye on aluminium electrodes (Merzouk *et al.*, 2011) while 98% decolourization efficiency was obtained for textile wastewater containing Orange II, a typical mono-azo acid dye (Chafi *et al.*, 2011). The inability to achieve colour removal efficiency above 80% in the electrocoagulation of azo-2-naphthol may be attributed to the presence polar substituents on the aromatic structure which induces solubility and does not enhance proper electrocoagulation, as compared to a non-polar group which could electrocoagulate easily due to immiscibility imposed by the non-polar group. Jain *et al.* (2009) also observed the difficulty of conventional methods to treat azo reactive dye, due to water solubility and polar compounds. Successful electrocoagulation in ethanol had been reported for 1,4-benzoquinone and 8-hydroxyquinoline (Mkpenie *et al.*, 2014). As observed, decolourization of PAN was faster in ethanol. The order of decolourization of PAN in the solvents could be given as follows: Ethanol>n-hexane>benzene>acetone. No trend could be identified to fit the order of decolourization given based on solvent properties such as dipole moment, dielectric constant, refractive index, solubility, hydrogen bond donor acidity, hydrogen bond acceptor basicity and polarizability/dipolarity parameter.

The Effective Colour removal efficiency (EC_R) of PAN was also evaluated. The EC_R is determined by dividing the percentage of colour removal by the time interval used (in this case, 15 min). This parameter affords the understanding of the amount of colour removed per time interval. The EC_R plot is shown in Fig. 3. The results show that varying amount of colour was removed within the same time frame. The first 15 min recorded the highest decolourization efficiency in all the solvents. Subsequently, colour removal was very minimal from 6.0% down to 1.0% in ethanol, 6.5-2.1% in n-hexane, 6.5-0.9% in benzene and 5.4-0.5% in acetone. This also shows that the amount of colour removed per 15 min interval was not constant throughout the electrocoagulation process.

Adsorption kinetic model: Adsorption is one of the major steps in electrocoagulation processes in which decolourization is believed to occur. The Al^{3+} and OH^- ions generated during the electrochemical reaction (anodic and cathodic reactions) react to form various monomeric and polymeric species and transform into $Al(OH)_3$ (Deb and Majumdar, 2013). The dye molecules

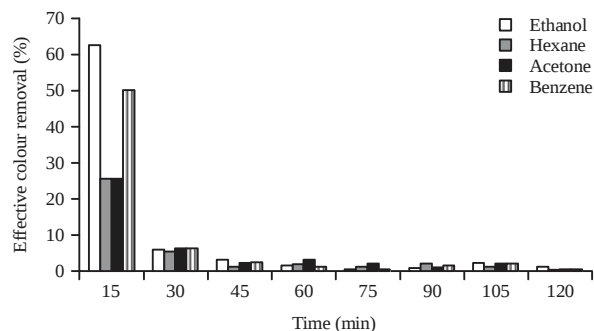


Fig. 3: Effective colour removal efficiency (EC_R) of PAN per 15 min interval in different solvents

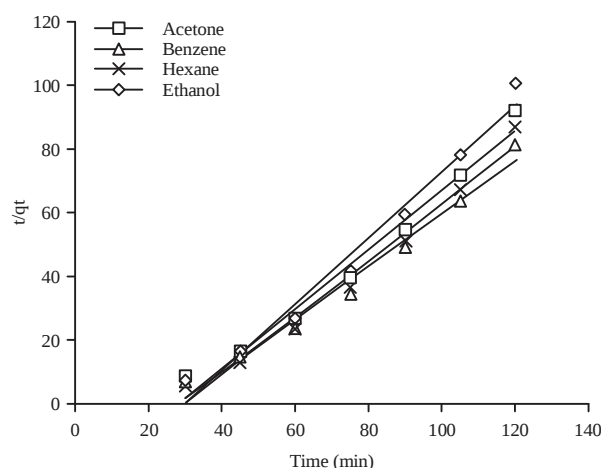


Fig. 4: A plot of t/q_t against t for electrocoagulation of PAN in different solvents (Pseudo-second order kinetic model)

present in the solution is adsorbed on the coagulants, $\text{Al}(\text{OH})_3$ (Holt *et al.*, 2005). Each combination of adsorbent and adsorbate has a distinct adsorbate-adsorbent interaction. It is necessary to determine the adsorption kinetic model that best fit the adsorption data so as to obtain the kinetic parameters involved in the adsorption process. Different adsorption kinetic models were investigated to determine the adsorption parameters. These include pseudo first-order kinetic model, pseudo second-order kinetic model, Elovich equation and intraparticle diffusion model. The experimental data obtained in this study fit the pseudo second-order kinetic model and the Elovich equation. The pseudo-second order kinetic model is given in Eq. 2 (Vasudevan *et al.*, 2009b):

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (2)$$

where, q_e and q_t are the amounts of adsorbate adsorbed (mg L^{-1}) at equilibrium and at contact time, t (min), respectively, while k_2 is the pseudo second-order rate constant (min g g^{-1}).

The plots of t/q_e against $t(\text{min})$ for the adsorption of PAN in ethanol, n-hexane, benzene and acetone, respectively, gave straight lines as shown in Fig. 4, which confirms that the adsorption of PAN in the used solvents obeyed the pseudo-second order kinetic model.

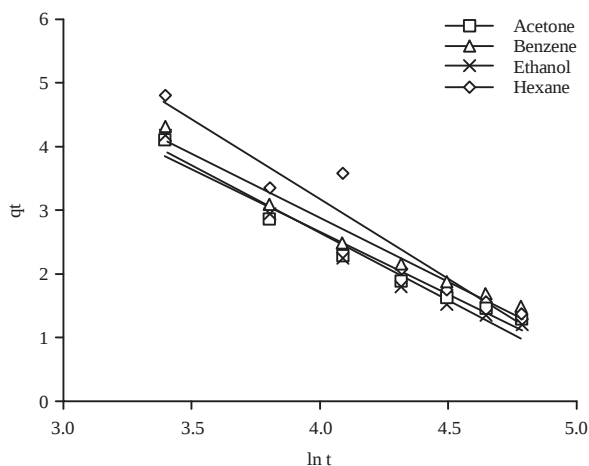


Fig. 5: A plot of q_t against $\ln t$ for electrocoagulation of PAN in different solvents (Elovich kinetic model)

Table 1: Kinetic parameter for the adsorption of PAN by different solvents at 27°C (Pseudo-second order kinetic model)

Pseudo-second order model and Parameters	Acetone	Benzene	Hexane	Ethanol
Experimental q_e (g g^{-1})	1.4722	1.6551	1.5601	1.3474
k_2 (min g g^{-1})	0.0287	0.0257	0.0266	0.0306
Calculated q_e (g g^{-1})	1.0691	1.2136	1.1162	0.9621
R^2	0.9804	0.9817	0.9791	0.9778

PAN: 1-Phenylazo-2-naphthol

The values of q_e and k_2 calculated from the slope and intercept of the plots are presented in Table 1 along with the corresponding values of the correlation coefficients. From the values in Table 1, it can be seen that the correlation coefficient values for the second order kinetic model in the four solvents studied were above 0.97 and also the calculated q_e values reasonably agree with the experimental q_e values. Similar pseudo-second order kinetic models have been reported in the adsorption of phosphate on Fe (III)/Cr (III) hydroxide (Namasivayam and Prathap, 2005), NO_3^- on iron (III) hydroxides adsorbents (Ghanim and Ajjam, 2013) and removal of cadmium ion on aluminium electrodes (Mansour *et al.*, 2013). Boron removal from synthetic wastewater also followed pseudo-second order model (Yilmaz *et al.*, 2008b).

The Elovich kinetic model is given in Eq. 3 (Ho and McKay, 1998):

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t \quad (3)$$

where, q_t (g g^{-1}) is the sorption capacity at time t , α is the initial sorption rate ($\text{g g}^{-1} \text{min}^{-1}$) and β is the desorption constant (g g^{-1}) during any one experiment. The linear plots of q_t against $\ln(t)$ (Fig. 5) also shows that the adsorption of PAN in the studied solvents obeyed the Elovich kinetic model.

CONCLUSION

The colour removal of azo 2-naphthol dye was achieved by electrocoagulation in various solvents including ethanol, acetone, n-hexane and benzene. The results showed that decolourization was faster in ethanol with an efficiency of 79% compared to other solvent investigated. Decolourization

of the dye followed the order: Ethanol>n-hexane>benzene>acetone. The percentage of effective colour removal (EC_R) showed that varying amount of the dye colour was removed in the time interval investigated. The highest decolourization efficiency was achieved during the first 15 min. The adsorption of PAN on aluminum hydroxide obeyed pseudo second-order and Elovich kinetic models.

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