



Electrocatalytic degradation and minimization of specific energy consumption of synthetic azo dye from wastewater by anodic oxidation process with an emphasis on enhancing economic efficiency and reaction mechanism

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ABSTRACT

This work focused on the knowledge-based methodology for the development of an electrochemical system, enabling simultaneous optimization of various operating parameters such as current density (j), initial dye concentration (C_0), NaCl concentration (C_N) for the mineralization of Reactive Violet 2 (RV-2) and Acid Brown 14 (AB-14) dye on the efficiency of removal, energy consumption (EC), Chemical Oxygen Demands (COD), apparent rate constants (k_{app}) and Electrical Energy per Order (E_{EO}) all of which have been examined. The relationship between k_{app} and E_{EO} is also discussed. The degradation efficiency and k_{app} always rising at higher j and lower C_0 and C_N while EC, E_{EO} , and operating cost increased at higher j , C_0 and C_N . On the other hand, The COD increased with decrease j , C_0 and higher C_N . Due to the strong formation of hydroxyl radicals from water discharge, the graphite electrode possesses a strong power of electro-generation rate and competitive wasting reactions of organic compounds. The results demonstrated that the relatively high dye removal, COD and low specific energy consumption are obtained simultaneously only if the various parameters are regulated to a plausible value j of 79 A m^{-2} , C_0 of 100 mg/L and C_N of 1 g/L within 60 min of electrolysis. The color removal efficiency is much faster for RV-2 compared to AB-14 due to the contribution of azo bond in the dye molecule. Also, the EC and k_{app} are higher for RV-2 than AB-14 while is lower in terms of E_{EO} and COD. A comprehensive reaction sequence of RV-2 and AB-14 mineralization involving all oxidation products was proposed. Formation and evolution of aromatic and aliphatic (short-chain carboxylic acids) intermediates during the treatment and a mineralization pathway is proposed. The estimated cost of operation for degradation at optimum conditions is calculated as 1.54 and 1.29 USD m^{-3}/g dye for complete degradation RV-2 and AB-14, respectively.

1. Introduction

One such major challenge is to keep the quality of water resources. The social, economic and environmental impact of water shortage needs to explore new techniques in the administration of water resources. One way to do this is to adapt preventive techniques to avoid discharging of untreated wastewater that including the dissolved organic pollutants to reuse in these industrial companies. The textile industry needs a large volume of water and around 90% of input is discharged into rivers, seas, etc., and it is one of the most contaminated water due to its intense color, high COD, large quantity of suspended solids, broadly fluctuating pH, high salt concentration and biotoxicity.

There are many techniques to decolorize the organic dye from color

wastewater such as; adsorption (Abd Elhafez et al., 2017; El Essawy et al., 2017), ion exchange (Elkady et al., 2016), precipitation (Zhu et al., 2007), Electrochemical methods (El-Ashtoukhy et al., 2017; Bassyouni et al., 2017); TiO_2 photo-catalysis (Hamad et al., 2015a, 2015b, 2016; Fathy et al., 2016), and biodegradation (Spasiano et al., 2016). Although the adsorption is effective in high removal efficiency of organic contaminants from an aqueous solution, the adsorbent materials suffer from several operational problems such as; separation inapplicable and high waste disposal treatment. So, many research groups all over the world explore and develop more efficient and economical methods which need minimum chemicals, electrode and energy consumptions and small installation space (Avlonitis et al., 2008).

As one of the widespread electrochemical methods, the anodic

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Nomenclature

A	Absorbance
APHA	American Public Health Association
M	Anode Material
AO	Anodic Oxidation
k_{app}	Apparent rate constant
COD	Chemical Oxygen Demand
AB-14	C.I. Acid Brown 14
RV-2	C.I. Reactive Violet 2

j	Current density
DC	Direct Current
EEO	Electrical Energy per Order
EC	Energy Consumption
IRS	Impeller Rotational Speed
C_o	Initial dye concentration
PTFE	Poly Tetra Fluoro Ethylene
C_N	NaCl concentration
R^2	Regression co-efficients
SHE	Standard Hydrogen Electrode

oxidation (AO) technique has been the subject of interest nowadays for the removal of the organic contaminants from effluents due to their high efficiency, environmental compatibility, amenability to automation, high energy efficiency, safety under mild conditions, non-requirement of large quantities of chemicals, no secondary pollutants and versatility (El-Ashtouky et al., 2017; Souza et al., 2015; Bassyouni et al., 2017). The electro-generation of high oxidation potential such as $\cdot\text{OH}$ and $\cdot\text{OCl}$ from AO on electrodes are counted highly efficient treatments (Bassyouni et al., 2017).

These methods are a great advantage of continuous in situ production of a highly reactive oxidizing agents such as $\cdot\text{OH}$, which are able to oxidize effectively all organic molecules into CO_2 (Oturan, 2014). It is able to react with an organic molecule to give dehydrogenated or hydroxylated derivatives to finally mineralization into CO_2 and inorganic ions in addition to water (Özcan et al., 2009).

AO is the method according to which organic dyes in water are oxidized by direct charge transfer on the anode (M) from hydroxyl radicals ($\text{M}(\cdot\text{OH})$) which generated from the oxidation of water (Eq. (1)) (Panizza and Cerisola, 2009).



One way of getting the best of some of these challenges in AO, is to explore and develop strategies for satisfying the high efficient energy utilization to reduce EC and operating costs coupled with high electrochemical oxidation efficiency for suitable market applications (Mu'azu et al., 2016). In order to accomplish that, graphite is used as anode and stainless steel as a cathode. It is some of the commercial, inexpensive, available, stable in solution, and non-active electrode materials which could significantly improve the cost-effectiveness with high oxygen over the potentials of AO (Lu et al., 2015).

The high efficiency must also be economically feasible with regard to its operational cost and initial capital. The economic aspects of AO process are not investigated well by research groups except by a few of them (Daneshvar et al., 2007). Energy consumption (EC), electrode consumption and the cost of materials of the anode and cathode are very important economically parameters in AO.

The major challenge is to study and compare the performance of anodic oxidation for the degradation of RV-2 and AB-14 dyes and therefore the reduction in color removal, EC and operation cost. The AO process is done in a batch reactor using stainless steel and graphite electrodes – abundant and cheapest electrode materials – as both cathode and anode, respectively. There are various ways which compared in terms of color removal efficiency and COD reduction, EC and cost analysis. Trials are carried out under comparable conditions so as to which dye has the highest oxidation power and is more cost effective by AO. Our study is focused on the growth of a powerful electrolytic system to decolorize dye solutions. To do this, the effect of j on the performance of AO is examined to better illustrate the role of generating oxidizing agents. Also, the influence of some operational parameters such as; C_o and C_N has been discussed. A general reaction sequence for RV-2 and AB-14 dyes mineralization involving all the products is finally proposed.

2. Materials and methods

2.1. Chemicals and reagents

The characteristics of commercially available RV-2 and AB-14 dyes were shown in Table S1. These two dyes were supplied by the Isma Dye Company, Kafr El Dawar, Egypt and were used as received. The solution pH was initially adjusted with analytical grade NaOH and HCl. NaCl was used as supporting electrolyte. All solutions were prepared with distilled water in order to prepare the desired concentration of dye stuff solution.

2.2. Experimental set-up

All experiments were carried out under a batch scale electrochemical oxidation unit of polluted wastewater using 2.5 L capacity in an undivided plexiglass cylindrical vessel with dimensions (35 cm height and 20 cm diameter). The vessel was equipped with four graphite rods act as anode of diameter 2 cm, fixed to the bottom of a container 90° from one another. Graphite was used as anode in view of the fact that O_2 over-potential was highly on graphite while Cl_2 over-potential is low. This would enhance the current efficiency of Cl_2 evolution. A cylindrical stainless steel screen cathode (mesh No. 10) was placed in front and behind the anode. Stainless steel was used as cathode in view of its stability in the solution. The distance between the anode and cathode was 2 cm. The solution was stirred by hydrogen gas evolved at the cathode. This sample setup displayed advantages such as; the uniformity of current and potential all over the anode surface as well as the screen cathode enhanced the mixing efficiency inside the cell and acts a turbulence promoter. The electrodes were attached to DC power supply (20 V, 10 A) with a voltage regulator and a multi-range ammeter connected in series. Samples were drawn at regular intervals of time during the experiment period using 10 ml pipette. All the experiments were performed at room temperature (25°C) and natural pH (7 for RV-2 and 6 for AB-14 dye). Experimental analysis was conducted at variable j, C_o and C_N .

2.3. Instruments and analytical procedures

The solution pH was evaluated using a pH meter (Jenway, UK). Samples were collected at regular time intervals, to stop the mineralization process, and filtered with $0.45\ \mu\text{m}$ PTFE filters purchased from Whatman before analysis. The decolorization of RV-2 and AB-14 dye solutions was followed by monitoring their absorbance (A) minimize at the maximum visible wavelength of $\lambda_{\text{max}} = 550$ and $450\ \text{nm}$, respectively using UV-vis spectrophotometer double beam (Labomed, U.S.A). The j of the electrochemical oxidation was estimated according to the Eq. (2) as follows:

$$j = I / (2 \times S) \quad (2)$$

Where j is the current density through the solutions (A) and S was the area of the electrode (m^2). The percentage of color removal in the treatment experiments is evaluated as follows:

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