Effect of initial pH value on the removal of reactive black dye from water by electrocoagulation (EC) method

Khalid S Hashim\textsuperscript{1,2}, Ameer H Hussein\textsuperscript{3}, Salah L Zubaidi\textsuperscript{4}, Patryk Kot\textsuperscript{1}, Layth Kraidi\textsuperscript{1}, Rafid Alkhaddar\textsuperscript{1}, Andy Shaw\textsuperscript{1}, Reham Alwash\textsuperscript{2}

\textsuperscript{1}Department of Civil Engineering, Liverpool John Moores University, UK. 
\textsuperscript{2}Department of Environmental Engineering/ University of Babylon, Iraq. 
\textsuperscript{3}Al-Mussaib Technical Institute /Al-Furat Al-Awsat Technical University, Iraq. 
\textsuperscript{4}Department of Civil Engineering, College of Engineering, University of Wasit, Iraq.

Email: k.s.hashim@2013.ljmu.ac.uk

Abstract. This study investigates the influence of the initial pH on the removal of reactive black 5 dye (RB5) from water using electrocoagulation (EC) reactor supplied with aluminium electrodes. The influence of the initial pH has been investigated by commencing several sets of continuous flow experiments at five different initial pH values (4, 5, 6, 7, and 8) keeping the current density, inter-electrodes distance, and the concentration of RB5 constant at 2 mA/cm\textsuperscript{2}, 4 mm, and 25 mg/L, respectively. The obtained results showed that the removal efficiency increased gradually as the initial pH increased from 4 to 6 to reach its maximum level (96%) at the neutral range of pH, then it decreased to 74% as the initial pH increased to 8. This change of the removal efficiency with the initial pH could be attributed to the predominant species of aluminium coagulants, where in alkaline and acidic conditions; the prevailing species have low adsorption capacity for pollutants. While, in the neutral range of pH, the predominant species have high adsorption capacity for pollutants.

Keywords: Reactive black dye; electrocoagulation; water.

1. Introduction
Textile industry is categorised as a major source for water pollution because it uses massive quantities of water, chemicals, and dyes during the production and treatment processes, which in turn generates huge volumes of polluted wastewater (Lotito \textit{et al.}, 2014). For example, it has been reported that the textile industry consumes 0.1-0.2 m\textsuperscript{3} of water to produce 1 kg of products, and uses 700000 tons of different dyes per year (De Jager \textit{et al.}, 2014; Sharma \textit{et al.}, 2016; Kraidi \textit{et al.}, 2019). Discharging this dyes containing wastewater into the water bodies threatens the human life, aquatic life, and the quality of water at large. For example, it produces carcinogenic agents due to the decomposition of azo dyes, it limits sun light penetration, and it decreases the concentration of the dissolved oxygen, that threatens the aquatic life (Gole and Gogate, 2014; Joshi and Mhatre, 2015; Santos and Boaventura, 2015; Hashim \textit{et al.}, 2018a). Therefore, a board rage of treatment methods were practised to treat the wastewater of textile industry, such as aerobic and anaerobic degradation, adsorption, and filtration (Mahmoud \textit{et al.}, 2012; Gole and Gogate, 2014; Kraidi \textit{et al.}, 2018; Zubaidi \textit{et al.}, 2018b). Unfortunately, most the conventional treatment methods are not efficient to remove azo dyes (Aşçı \textit{et al.}, 2015; Hayat \textit{et al.}, 2015; Zubaidi \textit{et al.}, 2018a). More worse, the produced water of some
treatment methods contains carcinogenic agents (Aravind et al., 2016). For example, the produced water from the biological reduction of azo dyes under anaerobic conditions contains aromatic amines, which categorised as carcinogens (Lourenço et al., 2015).

According to the relevant literature review, there is a serious need for efficient, affordable, and eco-friendly treatment method for the textile wastewater. Thus, a number of advanced treatment methods were recently practiced to treat the effluents of the textile industries, such as the nanofiltration (Ong et al., 2014), membrane bioreactor-ultrafiltration (De Jager et al., 2014), and electrocoagulation (Vidal et al., 2016; Hashim et al., 2018b; Hashim et al., 2018c; Hashim et al., 2017a; Hashim et al., 2017c; Hashim, 2017). Amongst these effective methods, electrocoagulation method (EC) received a big deal of interest during the last few decades due to its attractive advantages, such as it can treat different pollutants at the same time, it requires short treatment time, and it does not require chemical additives (Hashim et al., 2017b; Hashim et al., 2016a; Hashim et al., 2015a; Hashim et al., 2015b; Hashim et al., 2016b; Shaw et al., 2017). Additionally, the EC method produces small volumes of sludge in comparison with other treatment methods that greatly enhances its cost-effectiveness (Hashim et al., 2017a; Hashim et al., 2017c), because the produce sludge requires expensive treatment (Abdulredha et al., 2018; Abdulredha et al., 2017; Alattabi et al., 2017a; Alattabi et al., 2017b; Al-Jumeily et al., 2018).

In this context, the current study is devoted to explore the effect of a key operating parameter, the initial pH, on the performance of the EC units in terms of dye removal from water. In this study, RB5 was selected as the model dye because it is one of the widely used dyes in the textile industry, and its degradation generates toxic compounds (Chang et al., 2010). The molecular structure of RB5 is shown in Fig. 1.

![Molecular structure of RB-5 dye (Lucas and Peres, 2006).](image)

**Figure 1.** Molecular structure of RB-5 dye (Lucas and Peres, 2006).

### 2. Experimental work

#### 2.1. Material and methods

The azo dye, RB5, and other chemicals were supplied by Sigma-Aldrich UK and used as supplied. Stock solution, 100 mg/l, was prepared by dissolving the required RB5 in deionised water. Samples with lower RB5 concentrations were diluted from this stock solution. The initial pH of the diluted samples was adjusted to the desire level, in the range of 4 to 10, by using 1 M HCl or 1 M NaOH solutions. While the conductivity of these samples was adjusted to 0.32 µS/cm using the required quantity of NaCl. Both water conductivity and pH values were measured using Hanna meter (Model: HI 98130).

#### 2.2. EC unit

The electrolysis process has been carried out using a flow column EC unit. This unit consists of a plastic cylindrical container (25 cm in height and 10.5 cm in internal diameter) that is supplied with aluminium perforated discoid electrodes (10.4 cm in diameter and 0.1 cm in thickness), Fig. 2. These electrodes were stacked vertically within the cylindrical container with the plane of each electrode...
parallel and perpendicular to the direction of flow. Each electrode had the holes offset from the one above it to ensure that the water will flow in a convoluted path in order to increase mixing efficiency. These electrodes were held in the required position, inside the container, using PVC rods and spacers.

The EC unit was supplied with a peristaltic pump (Watson Marlow type, model: 504U) to circulate water inside the EC unit, and a rectifier (HQ Power; Model: PS 3010). It is noteworthy to mention that aluminium has been used as electrodes material due to its cost-effectiveness and low-oxidation potential (Hashim et al., 2017a; Hashim et al., 2017c).

2.3. Electrolysing process

The electrolysing process has been carried out by pumping the coloured water samples continuously, at a flow rate of 1000 mL/h, into the EC unit, and subjected to a constant current density of 2 mA/cm². The distance between electrodes and initial water temperature were kept constant, during the course of experiment, at 4mm and 20±1°C.

The removal of RB5 was measured by collecting 5 mL water samples at 5 min intervals during the course of experiment. The collected samples were filtered at 0.45 µm filter papers (Whatman filters); the residual RB5 concentration in the filtrate was measured using a spectrophotometer (Hach Lange DR 2800). RB5 removal efficiency (RE %) was determined as follows (Hashim et al., 2018c):

\[
RE\% = \frac{C_0 - C_t}{C_0} \times 100\%
\]

(1)

Where \( C_0 \) and \( C_t \) represent the initial and the measured concentrations of RB5, in mg/L, respectively.

3. Results and discussion

To investigate the influence of the change in the pH level on the removal of RB5 from water using the electrocoagulation technology, diluted water samples containing 25 mg/L of RB5 with different initial pHs (4 to 8) were electrolyzed for 60 min at flow rate of 1000 mL/h, current density of 2 mA/cm², distance between electrodes of 4 mm, and water conductivity of 0.32 mS/cm.

The obtained results, Fig. 3, showed that the removal of RB5 is not sensitive from the change of initial pH value within the slightly acidic and neutral environments. However, the removal of RB5 is very sensitive for high pH values. Where, it has been found that RB5 removal is inversely proportional to the level of pH (within the studied range of pH), where it has been noticed that found that the removal of RB5 decreased from about 99.7% to 98.6%, 94.8%, and 80.3 % as the initial pH increased from 4 to 5, 6, and 8, respectively.

This change in dye removal could be explained by the change in the amphoteric characteristics of aluminium hydroxide, where the predominant aluminium hydroxides in the neutral and slightly acidic environments is Al (OH)₃, which has high adsorption capacity. While, in alkaline environment, Al (OH)₄, which has low adsorption capacity, is predominant one (Emamjomeh and Sivakumar, 2009; Un et al., 2013). Therefore, it could be conclude from the results of the current work, that the initial

![Figure 2. The EC unit.](image-url)
pH of 5.0 is the best value to remove RB5 from water using aluminium-based EC unit. It is noteworthy to highlight that this conclusion is only applicable for the studied range of pH.

**Figure 3.** Influence of the initial pH on RB5 removal.

4. **Conclusions and recommendations**

The outcomes of the current investigation confirmed that the performance of EC technology, in terms of reactive black 5 dye from water, could be enhanced by providing a neutral pH environment. Additionally, it could be concluded that, for the studied type of EC units, there is no need to add chemicals to increase the pH of slightly acidic water as the aluminium-based EC units is not sensitive for the change in the pH value within the slightly acidic and neutral ranges. Finally, the authors recommend, according to the outcomes of the current study, to investigate the influence of low pH values (such as 3) on the removal of RB5 from water. Additionally, the influence of other parameters, such as the distance between electrodes, on RB5 should be investigated.

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