A short review on dyes removal from water and wastewaters

Hayfaa A. Mubarak\textsuperscript{1,*}, Marwa A. Kubba\textsuperscript{2}, K. Hashim\textsuperscript{3}, Abduljaleel Al-Janabi\textsuperscript{4}, Safaa K.H\textsuperscript{4}

\textsuperscript{1} Chemical Engineering Department, University of Babylon, Babel, Iraq
\textsuperscript{2} Al-Rasheed University College, Baghdad, Iraq
\textsuperscript{3} College of Environment, Al-Qasim Green University, Babylon, Iraq
\textsuperscript{4} Engineering Faculty, University of Babylon, Babel, Iraq

\* E-mail: haifaadnan_81@uobabylon.edu.iq

Abstract. Dyes are applied broadly in the textile industry to colour the products and give the products the final desire look. Additionally, dyes and colouring agents are used in a wide range of industries, like the papers production, packing, food and plastic industries. This means dyes and pigments represent an important pile in the global economy and civilisation. However, dyes used in this wide range of applications has a grave impact on human life due to the serious effects of the wasted dyes, in wastewater, on aquatic life, the chemistry of water and human health. Hence, the need for the removal of dyes of the produced wastewaters is an important step in any type of industry to avoid the mentioned impacts. The removal of dyes or colouring agents could be achieved in several ways, such as adsorption on active surfaces, electrolysis the colouring agent, application of oxidation chemicals, and filtration of coloured solution. This paper shortly reviews the removal of dyes from solutions by electro-coagulation method because this method is common nowadays, especially in developing countries. The results of this short review highlighted the electro-coagulation method is an effective and affordable method and suitable for the removal of dyes from water or wastewater. The EC method is also safe for the environment because there are no chemical additives in the treatment. Thus, this method could be a good option for developing countries.

1. Introduction

Dyes or colouring agents are substances or materials that chemically bond to the materials exposed to, and it is different from other chemicals that do not form chemical bonds with dyed or coloured materials, the latter called pigments [1]. The dyes could be classified according to:

1.1. Their sources
   a) Natural dyes: produced from natural materials, such as berries, jack fruits, tea leaves, turmeric, fungi, indigo, and animals
   b) Synthetic dyes: produced by mixing two or more chemicals, such as Alcian blue, Sulpho orange, Cresyl blue and reactive red

1.2. Their chemical structures
   a) Disperse dye, such as disperse yellow 3
   b) Reactive dye, such as reactive dye 5
   c) Solvent Dye, such as solvent yellow 32
d) Direct dye, such as direct red 12B

e) Acidic dye, such as red acid 8

f) Basic Dyes, such as Acid yellow 36

g) Vat Dyes, such as Vat Black 25

Generally, the dyeing process is done by adding the dye or the agent into water or other aqueous solutions, and then the produced solution will be applied to the products that could be fabrics, food products, packing materials or other materials [2-4].

The dyeing process is not a new human activity, where dyed flax fibres (dated to 36,000 BP) were discovered in a prehistoric cave in Georgia. In fact, the dyeing process was used during the Neolithic periods, when the people started dying their fabrics natural materials such as fruits, peels, animals and roots and leaves of plants, for example, the crimson kermes was produced during the ancient and medieval worlds, and it was considered as a luxurious type of dyes. Additionally, dyes that were produced using woods, indigo and saffron were found in ancient Asia and Europe. The good fact about natural dyes is their minor effects on the environment and public health [5-7].

Unfortunately, the production of natural dyes is not enough to meet the increasing demand for dyed products, such as fabrics, backing items and food, due to the patterns of modern life [7]. For example, the global population has increased by many folds during the last few decades [8-10], and the urban areas were expanded by many folds [11-15]. As a result, the need for dyed clothes [16], food [17], packing products [18] and other daily needs [19-22] has increased rapidly and hugely; therefore, synthetic dyes were produced and became the predominant type of dyes nowadays in all industries [23-25]. The first type of synthetic dyes, called mauve, was developed serendipitously by William H. Perkin in 1856, followed by a dramatic increase in the production and consumption of dyes and colouring agents, especially during and after the 21st century. Nowadays, the number of the synthetic dyes is more than 100,000 types, and the global production of these dyes is more than 700,000 tons/year. The average consumption of freshwater during the dyeing process is between 100 and 200 litters per 1 kg of fabrics depending on the colouring technology and type of fabrics.

This increase in the consumption of dyes was accompanied by a significant increase in the generation of dyed wastewaters that has the main role in the deterioration of water quality and the spread of serious diseases [26-28]. For instance, dyes could cause carcinogenic diseases due to their chemical compositions that contain toxic elements, besides the consumption of the dissolved oxygen in the aquatic environment, the limitation of the sunlight penetration in surface water, and poising the aquatic life [29-31].

Therefore, several treatment approaches were experienced to remove the residual dyes and colouring agents from wastewaters before discharging them to the sources of water, aerobic and anaerobic degradations [32], adsorptions [33], filtrations [34], and electrochemical treatments [35-37] are good examples of the nowadays used approaches for the removal of dyes from wastewaters. Not all mentioned methods are applicable in poor or developing countries because many advanced methods, such as membrane filtration, require high capital cost and well-trained staff to operate them, which can not be found in many developing countries [38-40]. Therefore, many developing countries focus on the use of locally available methods, which do not require high costs or trained staff to operate, such as the electrocoagulation method.

The Electrocoagulation method is adopted on a large scale in developing countries because it can be manufactured using affordable local materials. However, this method is also operated by not well-trained staff. Figure 1 shows the general EC system. Therefore, this paper highlights the concepts of the EC method and some of the recent studies about the use of this method in the treatment of water and wastewater.
2. Electro-coagulation method

2.1. Principles of the EC method

Electrocoagulation (EC) method is one of the electrochemical methods that depend on metallic electrodes to produce, in situ, coagulants to remove the pollutants [3].

Generally, the EC units are made from anode and cathode (could be more than one pair) connected to a DC power. The DC current motivates the anode to produce positively charged ions and the cathode to produce hydrogen gas. The ions travel towards the negative electrode (the cathode); during this travel, the negative ions react with the pollutants in the solution (they usually have a positive charge), forming a floc (Figure 2), the latter grow in size until it reaches a heavyweight that can not be carried by the solution anymore [1]. As a result, the heavy flocs will be removed from the solution via precipitation and collected from the bottom of the unit as sludge. Another removal scenario happens when the applied current is high that motivate the cathode to produce more hydrogen gas in the form of bubbles. Because

![Figure 1: The EC system.](image1)

![Figure 2: The mechanisms of the EC method.](image2)
the produced bubbles are lighter than the solution, they will move upward to leave the solution; during this travel, flocs will be attached to these bubbles and accumulated on the surface of the solution as foam, which will be scammed later using a metallic plate [5].

2.2. The reaction in the EC method

The reactions in the EC method depends on the type of electrodes. Because the most used metals in the EC system is iron and aluminium. Therefore, the reactions of the aluminium and iron are listed here as follows [23]:

2.2.1. Aluminium electrodes

At anodes:

\[ Al(s) \rightarrow Al^{3+} + 3e^- \]  

At cathodes:

\[ 2H_2O + 2e^- \rightarrow H_2(gas) + 2OH^- \]  

Then, Al speciated according to the following equations:

\[ Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+ \]  
\[ Al(OH)^{2+} + H_2O \rightarrow Al(OH)_{2}^+ + H^+ \]  
\[ Al(OH)_{2}^+ + H_2O \rightarrow Al(OH)_{3}^0 + H^+ \]  
\[ Al(OH)_{3}^0 + H_2O \rightarrow Al(OH)_{4}(aq) \]

2.2.2. Iron electrodes

The following reactions take place when iron (Fe) electrodes are used [41]:

At anodes:

\[ 4Fe(s) \rightarrow 4Fe^{2+} + 8e^- \]  
\[ 4Fe^{2+} + 10H_2O + O_2 \rightarrow 4Fe(OH)_3 + 8H^+ \]  

At cathodes:

\[ 8H^+ + 8e^- \rightarrow 4Fe(OH)_3 + 4H_2 \]

The overall reaction is:

\[ 4Fe + 10H_2O + O_2 \rightarrow 4Fe(OH)_3 + 4H_2 \]

2.3. Advantages of the EC method

The electro-coagulation method acquired wide attention because of many advantages, which can be summarised as follows [42-46]:

1- The flocs are generated because of the magnetic fields (positive and negative charges); therefore, there is no water captured inside the flocs, which means the expensive dewatering process. Thus, this feature makes the EC cost-effective method.
2- The EC method is run using DC power, which could be obtained from the sunlight, wind or dry batteries; therefore, the EC method could be portable and used in disasters.

3- The EC is easy to be operated without the need for trained staff.

4- The EC uses metallic electrodes to produce coagulants; therefore, no need to use chemicals in the treatment. Hence, the EC has no negative impacts on the environment.

These advantages are behind the wide use of the EC in the current time, especially in developing countries. However, some minor drawbacks of the EC method are the appearance of the inert layers on the aluminium electrodes and the lack of designs of the EC units.

2.4. Recent uses of the EC method in the treatment of water and wastewater

Table 1 summarises some of the recent studies about the use of the EC in the treatment of both water and wastewater.

| Dye                | Metal (electrodes) | Flow   | Operating parameters (optimum)                                                                 | Best removal | Authors                                                                 |
|-------------------|--------------------|--------|-----------------------------------------------------------------------------------------------|--------------|------------------------------------------------------------------------|
| Acid Orange II    | Granular activated carbon | Batch | Time = 30 min, cell voltage = 20V, air flow = 0.1m³/L, gap between electrodes = 14 mm.          | 87%          | [47]                                                                   |
| Acid Red 14       | Iron anode and steel cathode. | Batch | Time = 4 min, current density = 8 mA/cm², pH = 6-9, gap between electrodes = 4 mm.              | 83%          | Daneshvar, Sorkhabi and Kasiri [48]                                     |
| Eosin yellowish   | Mild steel         | Batch | Time = 15 min, current density = 1.61 mA/cm², Power consumption = 1.5 KWh/m³, pH = 6.8, gap between electrodes = 12 mm. | 97%          | Golder, Hridaya, Samanta and Ray [49]                                   |
| Acid yellow 36    | Iron               | Batch | Time = 6 min, current density = 12.78 mA/cm², pH = 8, gap between electrodes = 25 mm.          | 83%          | Kashefiasl, Khosravi, Marandi and Seyyedi [50]                          |
| Levafix orange E3 GA | Aluminium         | Batch | Time = 12 min, current density = 10 mA/cm², Power consumption = 35                             | 95%          | Koby, Demirbas, Can and                                                 |
| Dye Type                  | Material   | Process                | Conditions                                                                 | Efficiency (%) |
|--------------------------|------------|------------------------|-----------------------------------------------------------------------------|----------------|
| Red dye                  | Aluminium  | Continuous             | Time = 14 min, current density = 31.25 mA/cm², Power consumption = 3.2 kWh/kg dye, flow rate = 37.2 L/h, pH = 6-9, gap between electrodes = 11 mm. | 85%            |
| Reactive Black 5         | Iron       | Batch                  | Time = 5 min, current density = 4.575 mA/cm², Power consumption = 4.96 kWh/kg dye, pH = 5, gap between electrodes = 25 mm. | 98.8%          |
| Orange II                | Aluminium  | Continuous             | Time = 7.3 min, current density = 16 mA/cm², Power consumption = 3.2 kWh/kg dye, pH = 6.5, flow rate = 350 mL/min, gap between electrodes = 6 mm. | 94.5%          |
| Direct red 81            | Aluminium  | Batch                  | Time = 60 min, current density = 1.875 mA/cm², pH = 6, gap between electrodes = 15 mm, | 98%            |
| Reactive Red198          | Aluminium  | Batch                  | Time = 30 min, cell voltage = 20V, pH = 5.5, Power consumption = 1.303 kW/m³, gap between electrodes = 10 mm, | 98.6%          |
| Acid Yellow 220          | Aluminium  | Batch                  | Time = 7.5 min, current density = 4 mA/cm², pH = 5, Power consumption = 950 kWh/m³, gap between electrodes = 10 mm, | 97%            |
| Direct red 81            | Aluminium  | Continuous             | Time = 7.3 min, current density = 20 mA/cm², Power consumption = 90.2%        |                |
| Dye                  | Metal   | Treatment   | Conditions                                                                 | Percentage | Authors                                    |
|---------------------|---------|-------------|----------------------------------------------------------------------------|------------|--------------------------------------------|
| Methylene blue      | Iron    | Batch       | 3.2 kWh/kg dye, pH = 7.5, flow rate = 10 L/H, gap between electrodes = 10 mm. | 100%       | Lapicque and Leclerc [58]                  |
| Imperon violet KB   | Aluminium| Batch       | Time = 24 min, current density = 5 mA/cm², Power consumption = 3.8 kWh/m³, pH = 9, gap between electrodes = 10 mm. | 98.5%      | Alizeh, Ghaframani, Zarrabi and Hashemi [59] |
| Orange II dye       | Aluminium| Batch       | Time = 10 min, current density = 4 mA/cm², Power consumption = 4.66 kWh/m³, pH = 4.57, gap between electrodes = 10 mm. | More than 99% | Naje, Chelliapan, Zakaria and Abbas [60] |
| Acid orange 2       | Iron electrodes | Batch | pH of solution=7, the current density=2 mA/cm², gap between electrodes=0.5 cm, and treatment time=40 minutes. | 100%       | Jawad, Saddam, Adaami, Kareem, Abdulredha, Mubarak, Kot, Gkantou and AlKhayyat [62] |

3. Conclusion
The obtained results from this short review showed many facts about the EC method, which are:

1- The EC method is eco-friendly as it does depend on metallic plates to provide the coagulants instead of the additions of external chemicals; this means no secondary pollutants will be produced.
2- The operating cost of the EC method is low because it depends on the DC current that could be provided using sunlight, winds or dry batteries.
3- The flocs are generated because of the magnetic fields (positive and negative charges); therefore, there is no water captured inside the flocs, which means the expensive dewatering process. Thus, this feature makes the EC cost-effective method.
4- The EC is easy to be operated without the need for skilled staff.
5- Although the EC has many advantages, it has some drawbacks, such as the appearance of the inert layers on the aluminium electrodes and the lack of designs of the EC units.

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