Decolourization of dye-containing solutions by electrocoagulation

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In the electrocoagulation of Drimarene Discharge X-3LG® (R12S) and Samaron Yellow 4® (D05H), the effects of sacrificial anodes, initial pH, rate of stirring and concentrations of dyes on the fraction of decolourisation were tested. The experimental results revealed that the colour of dyes in the aqueous phase was effectively removed when iron and aluminium were used as sacrificial anodes and the concentration of the dyes was less than 100 p.p.m. When the concentration of R12S exceeded 300 p.p.m. iron was a more suitable sacrificial anode for electrocoagulation of R12S than aluminium. For the electrocoagulation of D05H in aqueous solution aluminium as a sacrificial anode was superior to iron when the concentration of D05H exceeded 300 p.p.m. Using iron as sacrificial anode, the residual concentration fraction of R12S was decreased below 10% when the initial concentration of R12S and charge passed were 500 p.p.m. and 120°C, respectively. The decolourization fraction of D05H was 99% when aluminium was used as sacrificial anode.

1. Introduction

The characteristics of wastewater from textile dyeing are high or low pH, high temperature and a high concentration of colouring material. The methods of treatment of dyeing waste water are biological treatment, chemical coagulation, activated carbon adsorption, ultrafiltration and ozonization [1, 2]. Although the cost of biological treatment of dyeing waste water is less than that of other methods, the smaller efficiency of decolourization using this treatment is due to the toxicity of the dissolved or suspended matter [3]. The colouring matter of dyeing waste water can be effectively removed by methods of chemical coagulation, adsorption on activated carbon, ultrafiltration and ozonization. The costs of adsorption, ultrafiltration and ozonization exceed that of chemical coagulation.

Secondary pollution may be caused by chemical substance added at a high concentration when chemical coagulation is used to treat dyeing waste water. Excessively added coagulants can be avoided by electrocoagulation. The coagulant in this case is generated by electrooxidation of a sacrificial anode. The characteristics of electrocoagulation are simple equipment and easy operation, brief reactive retention period, decreased or negligible equipment for adding chemicals and decreased amount of sludge [4].

Therefore electrocoagulation has been widely used to treat waters containing food and protein wastes [5, 6], oil wastes [7–10], synthetic detergent effluents [11], mine wastes [12] and heavy metal-containing solutions [13–17]. Electrocoagulation can also be used to remove phosphate [18, 19], for the defluoridation of water [20], and to treat potable water [21] and textile waste waters [2]. The fraction of decolourization of textile waste water exceeded 90% [2]. Iron and aluminium are generally used as sacrificial anodes. The matter in dyeing waste water is coagulated by iron(III) and aluminium hydrates or hydroxides produced from the sacrificial anodes. In this paper the electrocoagulation of Drimarene Discharge X-3LG® (reactive dye) and Samaron Yellow 4® (dispersive dye) in aqueous solutions with iron and aluminium as sacrificial anodes is described.

2. Experimental details

Iron and aluminium were washed with acetone to remove surface grease. The impurities on the surfaces of iron and aluminium were removed by dipping for 20 min in a solution which was prepared by mixing HCl aqueous solution (35.0%, 25 ml) and hexamethylenetetramine ((CH2)6N4) aqueous solution (2.8%, 50 ml). The dyes used in the experiments were R12S (Drimarene Discharge X-3GL®, Sandoz) and D05H (Samaron Yellow 4®, CAS). R12S is a reactive azo dye of structure

\[ \text{Dye} \rightarrow \text{NH} \rightarrow \text{Cl} \]

\[ \text{Cl} \]

\[ \text{Cl} \]

The hue and solubility of R12S are orange and >100 g dm\(^{-3}\), respectively. D05H is a dispersive azo dye. The hue of D05H is greenish yellow and D05H is suitable for polyester dyeing. The electrocoagulation current was 0.1 A and the sacrificial anode area

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was 5 cm$^2$. The distance between the sacrificial anode and the cathode was 1.5 cm. Electrocoagulation of dye-containing aqueous solution was carried out in a beaker (250 ml) with a magnetic stirrer to agitate the solution. At the beginning of a run, the desired concentration of dye in the aqueous solution (200 ml) was fed into the reactor, and the pH was adjusted to a desired value. The reaction was timed starting when the d.c. power supply was switched on. Samples were periodically taken from the reactor.

The precipitate in the sample was centrifuged at 700 r.p.m. for 10 min and the filtrate was obtained. Filtrate (1 ml) was added into buffer solution (pH 7, 4 ml) and the residual concentrations of R12S and D05H were determined according to optical adsorption at 400 and 500 nm, respectively.

3. Results and discussion

3.1. Aluminium as sacrificial anode

3.1.1. Effect of initial pH on the electrocoagulation of R12S. With aluminium as sacrificial anode, the decolourization of R12S was significantly affected by the initial pH under acidic conditions. As shown in Fig. 1, the residual fraction of R12S decreased to 70.1% over 14 min electrolysis with initial pH 2. In contrast, the residual fraction of R12S was decreased below 10% when the initial pH exceeded 3. Using aluminium as sacrificial anode, the dominant species are cationic monomers such as $\text{Al}^{3+}$ and $\text{Al(OH)}_2^+$ when a low initial pH is applied. The main mechanism for the coagulation of dyes is double-layer compression [22]. In this case, coagulant ($\text{Al}^{3+}$) at a high concentration is needed for effective removal of dyes in the aqueous solution [22]. Hence the coagulation efficiency to remove R12S at initial pH 2 was small within 14 min. When the pH was in the range 4.0–9.0, polymeric species (such as $\text{Al}_3\text{O}_4(\text{OH})_7^{2+}$) and precipitate $\text{Al(OH)}_3(\text{s})$ were formed. The dyes were effectively coagulated with the polymeric species and precipitated by the mechanisms of adsorption, charge neutralization and enmeshment [22]. Therefore, the residual fraction of R12S at the initial pH 2 was larger than that when the initial pH exceeded 3 in acidic solution.

When the initial pH was less than 9 in the alkaline region and the electrolysis period was 14 min, the residual fraction of R12S effectively decreased to 4.0% (Fig. 2). The residual fraction was 60.0% when the initial pH was increased to 10. As described above, the coagulation efficiency was promoted by the enmeshment of R12S with $\text{Al(OH)}_3(\text{s})$ in the neutral solution. When the initial pH increased to 10, the amount of $\text{Al(OH)}_3(\text{s})$ decreased and the concentration of monomeric anion $\text{Al(OH)}_4^-$ increased. The efficiency of electrocoagulation of R12S then decreased with initial pH larger than 10.

3.1.2. Effect of initial pH on the electrocoagulation of D05H. Increasing the initial pH from 2 to 3 and keeping the electrolysis period at 15 min resulted in decreased residual fraction of D05H in the aqueous solution from 87.1 to 1.9% as shown in Fig. 3. Upon further increase of the initial pH to 10 the residual fraction of D05H remained constant. The final solution pH increased from 2.4 to 8.7 and remained constant when the initial pH was increased from 2 to 10. These results revealed that the pH of electrocoagulation of D05H with aluminium as sacrificial anode was in the range 2.0 to 2.4 with initial pH 2.0. The absence of adsorption, charge neutralization and enmeshment in the electrocoagulation of D05H resulted in the small coagulation efficiency at the initial pH 2. With aluminium as sacrificial anode, the disperse dye D05H was