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Enhanced Electrocoagulation of Basic Green 4 and Acid Yellow 23 using Graphene Oxide as Adsorbent

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ABSTRACT

The main aim of this work is environmental protection by investigation of efficient electrocoagulation of dyes by aluminum electrode cell setup with graphene oxide adsorbent. The efficiency of dye removal was evaluated by the absorbance of the Basic green 4 and Acid yellow solution at 621 nm and 425 nm respectively. The operating conditions of electrocoagulation such as current density (10-30 mA/cm²), electrolysis time (5-20 minutes), pH of the medium (2 to 10), stirring speed (150-350 rpm), inter electrode distance (0.8 to 2 cm) and NaCl concentration (1.5-5.0 g/L) were optimized and found to be 30 mA/cm², 20 min, pH 8.0, 300 rpm, 2 cm and 5 g/L respectively. The results showed that the dye removal efficiency was enhanced to 99% by the addition of graphene oxide. The result of the present research work can be useful for an efficient large-scale treatment of industrial effluents contaminated with Basic green 4 and Acid yellow 23.

1. Introduction

Environmental contaminants are common in industrial effluents due their manufacturing process, raw material and product. The pollutants are harmful and provide negative impacts to environment and human health due its toxic nature [1-4]. Heavy metal contamination/domination in land and water is found due to lack of corporate socially responsible and very low attention in environmental protection. As many chromium is naturally available in earth crust and its adverse effects for human health [5-8].

There is large amount of studies on the usage of electro-coagulation method. Among this treatment of chemical materials mechanical polishing wastewater from a semiconductor plant by electrocoagulation was explored beneficially. Sludge settling is the notable evidence for these studies with respect to sludge settling velocities after electrocoagulation by velocity simulations [9]. The hybrid process of electrolysis/electrocoagulation and zero-valent iron activated persulfate oxidation showed a significant synergetic effect in attractive municipal sludge dewater ability, and has the potential for enhancing industrial sludge dewater ability. The optimal dewatering conditions for municipal sludge were voltage 40 V and 4.15 g/L Na₂S₂O₈ when zero-valent iron induced electrodes were applied [10].

The key area of some previous workers was the elimination of heavy metal ions viz. Cu²⁺, Cr³⁺, Ni²⁺ and Zn²⁺, from metal plating wastewater using electrocoagulation technique [12]. They studied to eliminate hexavalent chromium and COD from wastewater by electrocoagulation. The experiments were carried out in an electrochemical reactor using iron sacrificial electrode [13].

The variations of some characteristics of the mixed liquor solution in wastewater treatment plants under the effect of electrocoagulation (EC) technique has also been reported. Unlike conventional methods, electrocoagulation was performed using cylindrical perforated iron electrodes to achieve a good distribution of the applied DC field onto the municipal wastewater. Nanoparticles synthesis from various route is emerging trend now a days, viz., green synthesis, chemical synthesis, physical syntheses. In ECR treated sludge's have some special features due to its unique morphological behavior [14-16]

Electrocoagulation is an electrochemical technique for treating wastewater using electricity instead of expensive chemical reagents. This technique has several advantages as compared to conventional methods

in terms of use of simple equipment, ease of operation, less treatment time, reduction or absence of chemicals addition. Moreover, an electrocoagulation process provides rapid sedimentation of electro generated flocks and a less amount of sludge production. Electrocoagulation has the advantage of removing the smallest colloidal particles compared with traditional flocculation-coagulation, such charged particles have a greater probability of being coagulated and destabilized because of the electric field that sets them in motion.

2. Experimental Methods

2.1 Equipment Setup

The electrochemical reactor system is as shown in Fig. 1. The ECF unit mainly consists of aluminum sheet as cathode and anode and a DC power supply. Working volume of the electrochemical reactor was around 0.25 L, inter electrode distance was 1 cm and the effective electrodes area was approximately 18 cm². A digital DC source (PICO, 0-30V 0-5A CVCC MODE) was used to supply the power to the ECF system. A magnetic stirrer was used to provide thorough mixing to the solution.



Fig. 1 Electrochemical reactor setup

2.2 Choice of Electrode Material

Aluminium (Al) is one of metals which widely used in various industries such as construction, transportation, electrical component and packaging. Several thousand tons of Al is used every year, and in the meantime, a lot of Al scrap is also generated. The Al scrap can be reused as secondary Al in industrial sectors after melting. Thus, aluminium is used as electrodes for this electrocoagulation process.

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2.3 Experimental Method

The Basic green 4 and Acid yellow 23 dye solution of 0.25 L and concentration of 100 ppm was added separately to the electrocoagulation reactor system. Then the operating factors of current density (10-30 mA/cm²), electrolysis time (5-20 minutes), pH of the medium (2 to 10), stirring speed (150-350 rpm), inter electrode distance (0.8 to 2 cm) and NaCl concentration (1.5-5.0 g/L) were varied. The temperature was maintained at 25 °C. At the end of experiments, the solution was centrifuged to remove the sludge. The solution was centrifuged and the absorbance was measured by UV-Visible spectrophotometer (Deep Vision UV-VIS Single Beam spectrophotometer) at 621 nm and 425 nm respectively for Basic green 4 and Acid yellow 23. The efficiency of dye removal was calculated using Eqs.(1),

$$\text{Efficiency (\%)} = 1 - (\text{final absorbance} / \text{initial absorbance}) \times 100 \quad \text{-----(1)}$$

3. Results and Discussion

3.1 Effect of Current Density

The initial absorbance spectra of Basic green 4 and Acid yellow 23 solutions presented the highest peak at 621 nm and 425 nm respectively, thus the reduction in intensity of the highest peak was referred as the dye removal efficiency. After treatment by EC the absorbance was measured at 621 nm for Basic green 4 and 425 nm for Acid yellow 23 using a UV-VIS spectrophotometer. The dye removal efficiency was increased by increasing current densities as linear relation (Fig. 2). This is because a large amount of aluminium hydroxide particles (i.e., Al(OH)₃) are generated at the higher current densities [17-19]. The aluminium hydroxide particles act as coagulant to destabilize the colloid particles of dye, and remove the destabilized dye from the water. The results were shown in the Table 1.

Table 1 Effect of current density for Basic green 4 and Acid yellow

Current density (mA/cm ²)	Basic green 4		Acid yellow 23	
	Absorbance	Efficiency (%)	Absorbance	Efficiency (%)
0	0.8	-	1.2	-
10	0.3	63	0.5	58
20	0.05	83	0.1	80
30	0.004	92	0.01	90

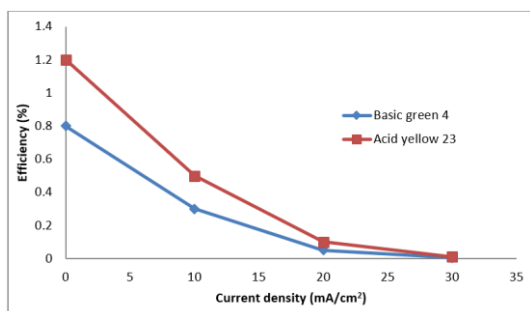


Fig. 2 Comparison chart of effectiveness of current density for dyes

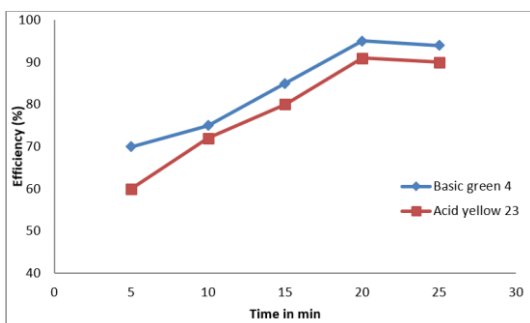


Fig. 3 Effect of electrolysis time dyes on dye removal efficiency

3.2 Effect of Electrolysis Time

At various electrolysis times, the dye removal efficiency was increased by increasing electrolysis times as exponential relation. The efficiency was sharply increased to 65% at 5 minutes, and continuously increased at the longer electrolysis times. The efficiency reached 75% at 10 minutes, then slightly increased to 84% at 15 minutes and reached its maximum of 95% at 20 minutes (Fig. 3). The longer electrolysis times causes high sludge

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formation and prevents the chemical interaction during coagulation process and increase the energy consumption.

3.3 Effect of pH

The nature and magnitude of total charge on metal hydroxide and dye molecules are pH dependent. Therefore, pH has a significant influence on the dye removal efficiency during electro-coagulation. Due to different organic structures of substrate (dye) and electrode materials, the effect of pH on anodic oxidation process becomes ambiguous. Plots of percentage dye removal efficiency as a function of pH at 20 minutes using Al as sacrificial anode was as shown (Fig. 4). It can be observed that at pH 8.0 the dye removal efficiency was maximum. The lower rate of color removal below optimum pH may be due to the availability of fewer hydroxyl (-OH) ions required for the formation of metal hydroxides flocculants. Further, on raising pH above the optimum value the color removal rate falls owing to diminishing positive charge at tertiary nitrogen of malachite green molecules, minimizing its interaction with the metal ions released by anodic oxidation and hence the lower rate of flocculation.

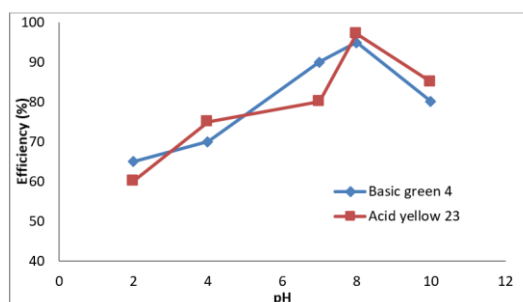


Fig. 4 Effect of pH on dye removal efficiency

3.4 Effect of NaCl Concentration

Sodium chloride played a vital role as conductive medium for electrocoagulation, and without NaCl addition the chemical reaction does not take place in the EC system. Concentration of NaCl was varied from 1 g/L to 5 g/L, the dye removal efficiency of Basic green 4 and acid yellow 23 was in the range between 80-97% (Fig. 5). During the reaction, efficiency increases with increase in the concentration of NaCl. It accelerates the reaction efficiency and at the end of experiments, the water became clear and colorless after sludge removal.

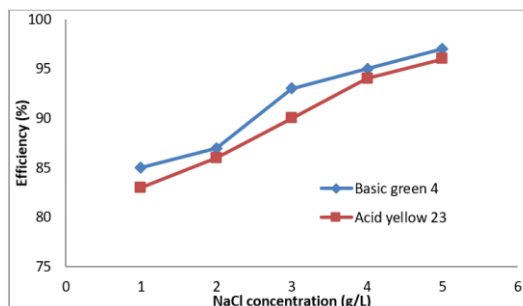


Fig. 5 Effect of NaCl concentration on dye removal efficiency

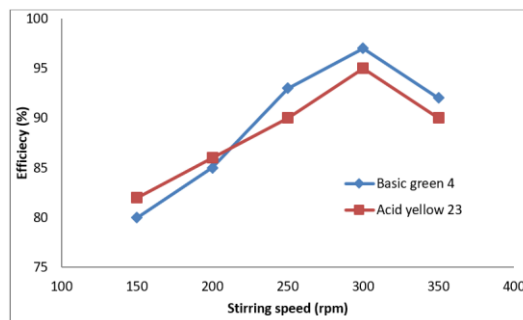


Fig. 6 Effect of stirring speed dyes removal efficiency

3.5 Effect of Stirring Speed

Influences of stirring speed on color removal efficiency of the dyes were studied by varying the stirring speed from 150 to 350 rpm. Till 300 rpm, notable and improved efficiency occurred with respect to stirring speed (Fig. 6). After 300 rpm the efficiency was decreased because of the aggregation of colloidal particles into flocs which could hinder the removal

process and has negative effect on colour removal. The main function of stirring is to ensure a good homogenization of the mixture dispersing the coagulant species in the reactor, thus causing efficient contact between the neutralized molecules and destabilized colloids and the cationic metallic species.

3.6 Effect of Inter Electrode Distance

In electrocoagulation process, electrochemical cell inter electrode distance regulates the reaction. To study the influence of electrode distance on colour removal efficiency, the distance between the electrodes was varied between 0.8 and 2.5 cm. The results obtained with the EC treatment shown in the Fig. 7. The removal efficiency is relatively low when the electrode distance was small and obtained its optimum at 2 cm (Fig. 7).

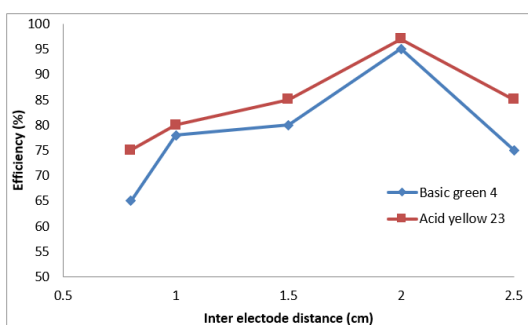


Fig. 7 Effect of electrode distance on dyes removal

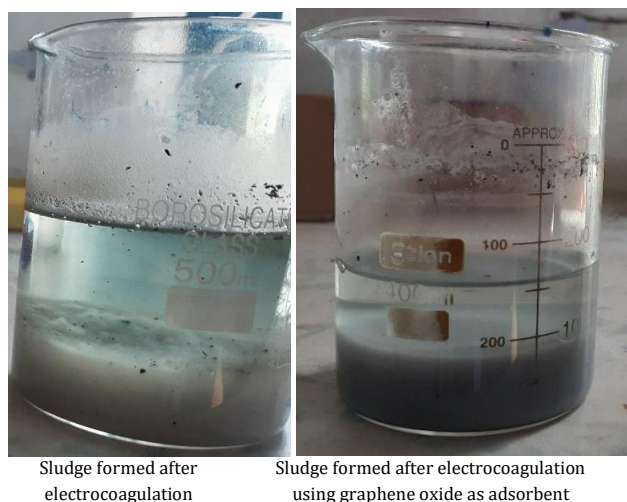


Fig. 8 Basic green 4 sludges with and without graphene oxide adsorbent

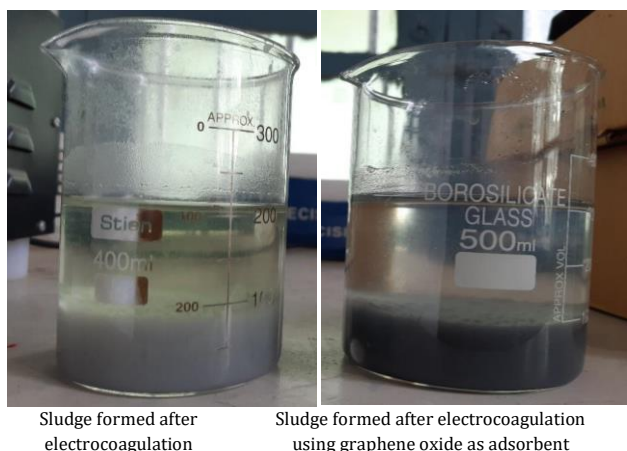


Fig. 9 Acid yellow 23 dye sludge's with and without graphene oxide adsorbent

3.7 Enhanced Activity of Electrocoagulation using Graphene Oxide

While optimization of all experiment results, 0.01 g of grapheme oxide was added with the dye solution of 0.25 L and concentration of 100 ppm in electrocoagulation reactor system. The operating factors were optimized and fixed as current density 30 mA/cm², electrolysis time 20 minutes, pH of the medium pH 8.0, stirring speed 300 rpm, inter electrode

distance 2 cm and NaCl concentration 5.0 g/L. The temperature was maintained at 25 °C. At the end of experiment, the surface of Al electrodes was slightly decayed to produce the aluminium ions and form the coagulant as aluminium hydroxide. Further, the sludge of aluminium hydroxide particles and dye were observed at the bottom, as shown in Figs. 8 and 9. However, the water became clear and colourless after sludge removal. Both dyes in EC reaction sludges are effectively settled in bottom, but slight improvement observed while using graphene adsorbent.

3.8 ECR Treated Sludge Analysis

The water removed sludges were packed in air tight containers and the parameters such as pH, TSS, COD, BOD of the samples after electrocoagulation were analyzed in Global lab and consultancy services, Salem, Tamil Nadu and the reports were given in Table 2. The sludge mainly contains aluminium hydroxide which was removed by centrifugation and dehydrated using water-miscible non-aqueous solvent like ethanol and formed into amorphous powder as shown in the Fig. 10. This powder can be used in the preparation of many fields like Fire retardant powders, zeolites, activated alumina and calcinated alumina.



Fig. 10 ECR treated aluminum hydroxide sludge from dyes after drying

Table 2 Test reports of slurries and centrifuges supernatant liquids

S. No.	Test Parameters	Test Method	Blue Green 4		Acid Yellow 23	
			Slurry mixture	Colourless liquor	Slurry mixture	Colourless liquor
1	pH	IS 3025 PART 11	6.69	6.74	6.31	6.69
2	TSS, mg/L	IS 3025 PART 17	54	42	58	54
3	BOD, mg/L	IS 3025 PART 44	250	120	261	215
4	COD, mg/L	IS 3025 PART 58	760	360	698	371

Table 2 reveals the test reports of blue green 4 slurry mixture found with higher amount of COD and BOD values and shows colourless water after Blue green 4 dye removal. It was looking good, no significance coloring after sludge deposition. Similarly, the transformations in before and after sludge depositing in Acid yellow 23 dye, extreme COD, BOD changes in ECR activity of sludge. The test reports reflect and proves the effectiveness of the electrocoagulation reaction.

3.9 AFM Analysis

Nanoparticles or tiny sized particles are measuring using this instrument. Fig. 11 shows the morphology of the Basic green 4 dye, it looks like floating sea morphology. It may due to the pollutants present in the dye solution. Fig. 12 represents the image of Acid yellow 23 dye, it also presents under micro region. Mixture of particles in dye solution makes the solution dense as well as unsmooth. Figs. 13 and 14 shows the AFM images of colorless water solution of Basic green 4 and Acid yellow 23 respectively. Particles are in nano region; it is due to higher agitation of mixture in ECR setup. Moreover, particles surfaces were clear, smooth and no notable defects, it proves the effectiveness of the reaction.

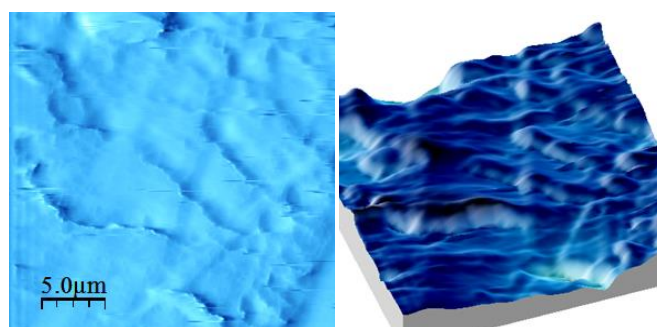


Fig. 11 Morphological images of Basic green 4 dye solution

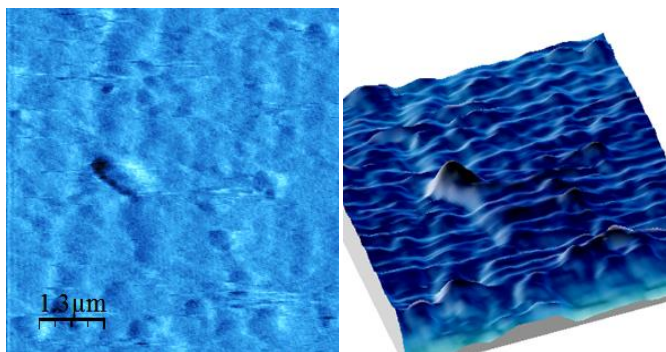


Fig. 12 Morphological images Acid yellow 23 dye solution

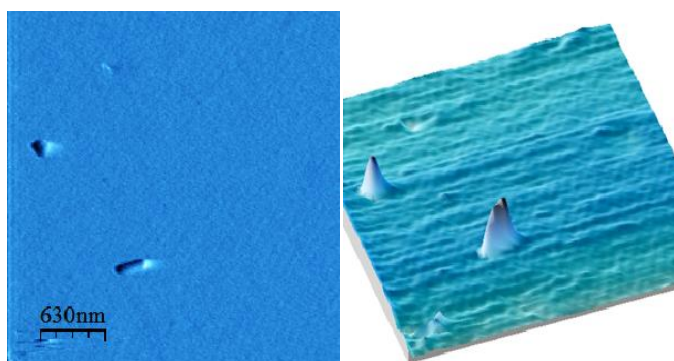


Fig. 13 Morphological images Acid yellow 23 dye solution

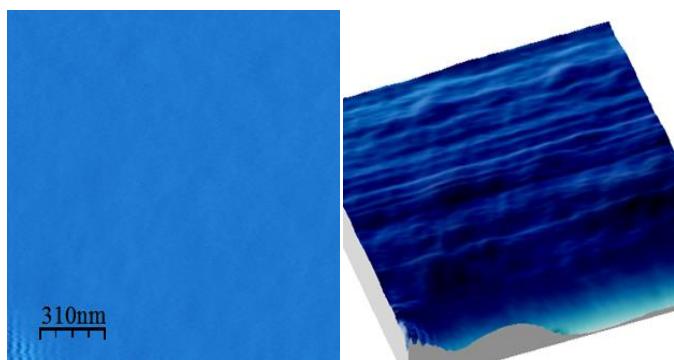


Fig. 14 Morphological images Acid yellow 23 dye solution

4. Conclusion

From the outcome it has been concluded that the electrocoagulation reaction (ECR) was effectively used for the removal of dyes viz., Basic green 4 and Acid yellow 23. Undeniably, the optimum operating circumstances was determined as current density 30 mA/cm², electrolysis time 20 minutes, pH 8, stirring speed 300 rpm, inter electrode distance 2 cm and NaCl concentration 5.0 g/L were optimized to produce environment beneficial products. ECR based these optimum parameters can provide the environmental protection for the society.

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