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REMOVAL OF MALACHITE GREEN DYE FROM AQUEOUS SOLUTION BY ELECTROCOAGULATION WITH STAINLESS STEEL ELECTRODES

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ABSTRACT

In order to see the influence of different variable parameters (inter electrode distance, initial conductivity, time of electrolysis, pH of the solution and initial concentration of dye) and the efficiency to remove Malachite Green dye (Basic Green 4) from aqueous solution, electrocoagulation (EC) method was employed. An electrocoagulation set up was designed using stainless steel as an electrode. Then different parameters were studied using known concentration (150 mg/l) and initial COD (256 mg/l) of prepared dye contaminated synthetic solution in a batch mode. The results showed that a very high de decolorization of 99.50% and the reduction of COD upto 85.71 % achieved with in 20 min electrolysis time, 1 cm of inter electrode distance and initial conductivity (1.5 mS) at pH 8. This confirms the efficient removal of dye.

KEYWORDS: Dyes, Electrocoagulation, Malachite Green Dye, COD, Color removal.

INTRODUCTION

Dyeing and finishing are the two chief processes usually applied in most of the textile industries generates great amount of wastewater contains strong color, dissolved and suspended solids and high chemical oxygen demand (COD) concentration. The disposal of these dye wastewaters causes a great problem for the industry as well as a threat to the environment. The majority of synthetic dyes are noxious substances to human and aquatic life. In textile industries, due to low dye fixing efficiency of 60–90% on textile fibers, large amounts of unfixed dyes are released in effluents. In the estimates around 1–15% of the dye is released into wastewaters during dyeing and finishing processes *(Daneshvar et al, 2006)*. The removal of dyes from colored effluent particularly from textile industries is one of the major environmental concerns these days and it is necessary to treat dye wastewater before discharged into water *(Tyagi et al., 2015).*

Malachite green (Basic Green 4 Dye) is one of the widely used dye which is used for dyeing silk, wool, leather, jute, and cotton, as a biological stain, as an acid-base indicator, as a dye for paper these vast use of Malachite Green result in its release to the environment through various waste streams. There are many harmful effect of this dye to environment and the human health such as when it is heated to decomposition it emits very toxic fumes of /nitrogen oxide and hydrogen chloride. Ingestion causes diarrhea and abdominal pain. Injurious to eyes, can cause bilateral blindness due to corneal problem *(Singh et al., 2013).* Hence, Malachite green was chosen for this research work

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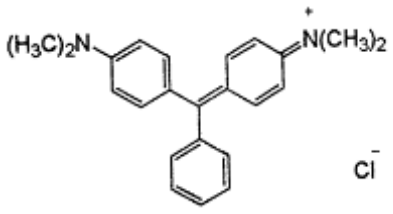

Fig.1.0 Molecular structure of Malachite Green Dye, (Source: Dye|World dye variety, Basic Green 4, 2015)

There are various techniques employed worldwide for removal of dyes from wastewater. The majority of these conventional methods is becoming inadequate and inefficient; because of the dye effluent contain mainly complex

aromatic molecular structures. Dye compounds are usually made to resist fading on exposure to soap; water, light and this make them more stable against biodegradation *(Carneiro et al., 2005)*. There are number of physical, chemical and biological treatment processes employed for the treatment of dye wastewater like Adsorption *(ELayazi et al, 2014)*, Chemical coagulation, Electrochemical treatments, Advanced oxidation processes (AOPs) including treatment with oxidizing agents, such as hydrogen peroxide (Fenton), ozone, UV light or their combinations, biological treatments such as anaerobic process, oxidation ponds, trickling filters, activated sludge process, etc. Physical methods such as Adsorption, Coagulation usually need additional chemicals which cause secondary pollution and a huge volume of sludge *(Vinodha and Jegathambal, 2012)*. In these processes there is excess generation of huge amounts of sludge and increases the total dissolved solids in the effluent. In biological treatment the microorganisms such as algae, fungi, bacteria, and yeasts are capable to degrade certain type of dyes. But, the application of biological treatment is limited as they requires a large land area, has sensitivity toward toxicity of certain chemicals and treatment time is very high. Further, in general some dyes are toxic and are not easily biodegraded by biological process *(Khandegar and. Saroha, 2013).* The sensitivity of biological treatment processes and secondary pollution caused by conventional chemical methods make advance treatment processes more popular.

Electrocoagulation is suitable methods for the treatment of such complex wastewater. It depends upon the electrochemical dissolution of sacrificial metal electrodes (usually iron or aluminium) into soluble or insoluble species that boost the coagulation, the adsorption or the precipitation of soluble or colloidal pollutants *(Holt, 2005).* A direct current supply is applied to metal electrodes immersed in wastewater. The flow of electrical current helps in the dissolution of metal electrodes into the solution. The dissolved metal ion, forms wide ranges of coagulated species and metal hydroxides at appropriate pH. It helps in destabilize and aggregate the suspended particles or adsorb and precipitate dissolved contaminants *(Arslan-Alaton et al., 2001, Murthy & Parmar 2011, Mollah et al., 2004, El-Ashtoukhy & Amin 2010*.)

The diverse reactions occurring at anode, cathode and solution are prescribed below *Irdemez et al., 2006), (Yildiz et al., 2007), (Wei et al., 2012* :

- At the cathode:
  $3H_2O + 3e^- \rightarrow 1.5\ H_2(g) + 3OH^-$ (1)
- At the anode:
  $4Fe(s) \rightarrow 4Fe_2^+(aq) + 8e^-$ (2)
- And with dissolved oxygen in solution:
  $4Fe_2^+(aq) + 10H_2O(l) + O_2(g) \rightarrow 4Fe(OH)_3 + 8H^+(aq)$ (3)
- Overall reaction:
  $4Fe(s) + 10H_2O(l) + O_2(g) \rightarrow 4Fe(OH)_3(s) + 4H_2(g)$ (4)

During electro-coagulation due to the change in pH various kind of monomeric and polymeric iron species formed are mentioned as follows:

$Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5OH^{2+}$, $Fe(H_2O)_4(OH)_2^+$, $Fe_2(H_2O)_8(OH)_2^{4+}$, $Fe_2(H_2O)_6(OH)_4^{2+}$ and $Fe(OH)^{4-}$ *(Sengil & Ozacar 2009).*

In this study, the removal of Malachite Green dye from a synthetic wastewater was investigated using electrocoagulation by Stainless steel electrodes and effect of various operating parameters like inter electrode distance initial conductivity, electrolysis time, initial pH and initial concentration of dye involved in electrocoagulation where examined and optimized. The optimum values of these parameters were primarily determined on the basis of color and COD removal.

MATERIALS AND METHODS

Materials and Methods

From the detailed literature review the Lab Scale Electrocoagulation reactor has been designed. Electro-coagulation set up has an internal dimension of 12.5 × 8 × 9 (L×B×H) cm, having a volume of 900ml with working volume 500ml. It is fabricated with 4mm thick acrylic sheet. It has been decided to use stainless steel as electrode materials. Each electrode has a wetted surface area of 94 cm^2. Magnetic stirrer is will be used for proper agitation. To find the removal efficiency of color and COD with stainless steel electrode, initially the applied voltage was kept constant at 15 V, initial pH and conductivity of the solution was kept at 7 and 1 m/S respectively and the time of electrolysis was 20 min.

The dye solution was prepared by dissolving Malachite Green (Basic Green 4) in distilled water. The dye was purchased from Oswal Udhyog, Mumbai, India was used as such without further purification. For the preparation of sample 150 mg of the dye was dissolved per liter of distilled water which is close to real industrial dye effluent.

Table 1.0 Initial characteristics of synthetic solution at various concentration of Malachite Green Dye

Concentration of Dye	COD	pH	Conductivity
50	64	7.70	0.10
100	192	7.58	0.26
150	256	8.14	0.41
200	25	8.25	0.70
250	320	9.25	0.75
300	320	9.30	0.79

Chemical Analysis

Intensity of color is measure based on various absorbance λ (nm) which are characteristics wavelength of various color. A UV-VIS Doublebeam spectrophotometer (Systronics AU-2701) was employed to measure the maximum absorption wavelengths for Malachite Green dye. Maximum absorption of Malachite Green dye used in experiments was λ_{max} 617 nm

Color removal efficiency can be determined from the formula, Color removal efficiency (%) =100x $\frac{(C0-C)}{C0}$

Where, C_0 and C are the concentrations of dye before and after electrocoagulation respectively.

COD was measured by closed reflux method as given in Standard Methods for the Examination of Water and Wastewater Analysis Manual 20th Edition (APHA, 1998) by suitable dilution of wastewater in distilled water if needed.

RESULTS AND DISCUSSION

Effect of Inter electrode distance on Color and COD removal

Inter electrode distance have great importance in EC process. Initially the inter electrode distance was varied from 1 cm to 4 cm. In the experiments when the distance was increased, the efficiency of the system decreases due to the lacking of metal ions for adequate coagulation. This may be due to the electrostatic attraction between the ions generated from the electrodes. For a constant current density, the generation of ions decreases on increasing the electrode distance due to the resistance between the electrodes increases. Due to this, the movement of ions becomes slower and the ions get enough time and space to flocs formation needed for settling of the impurities *(Khandegar and Saroha, 2012)*. It has been observed that keeping distance less than 1 cause problem in functioning and may cause short-circuiting in the set up and very high spacing causes the lesser dissolution of metal ions into the wastewater which reduce the effectiveness of the process. The maximum efficiency was obtained at 1cm for both color and COD which was the minimum amongst the selected range of distance. Fig. 2 shows the percentage removal of color and COD on varying inter electrode distance.

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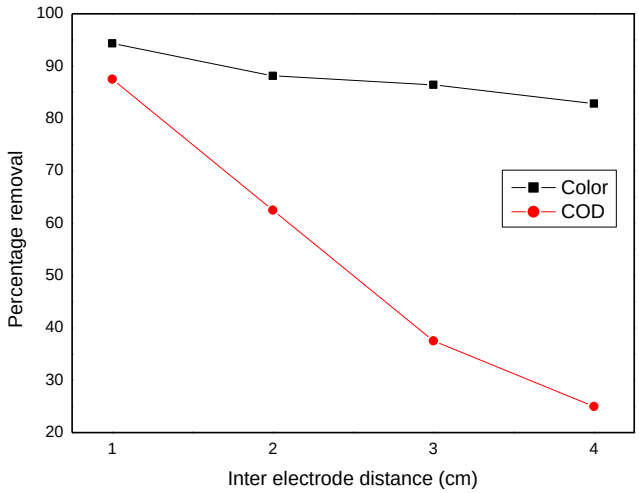

Fig. 2 Percentage removal of color and COD on varying spacing between the electrodes

Effect of Initial Conductivity on Color and COD removal

It is known from literature that conductivity is the function of the concentration of electrolyte in the solution. Therefore in the experiments conductivity was adjusted by adding Sodium Chloride (NaCl) in to the solution. The conductivity of the synthetically prepared solution was kept between 0.5, 1.0, 1.5, 2.0 and 2.5 mS during the experiments. The optimum conductivity for the electrocoagulation process in color and COD removal was found to be 1.5 mS and was selected for further experiments. Fig. 3 shows the percentage removal of color and COD on varying initial conductivity. The removal efficiency was found to increase with an increase in the conductivity. There should be an optimum conductivity of the solution in the electrocoagulation process because at higher conductivity due to high concentration of NaCl, improper dissolution of metals takes place causing troubled interaction between coagulant and particles. At high conductivity there is more production of sludge which is also responsible for the decrease in the removal efficiency of COD and color from the wastewater *(Arslan-Alaton et al., 2008).*

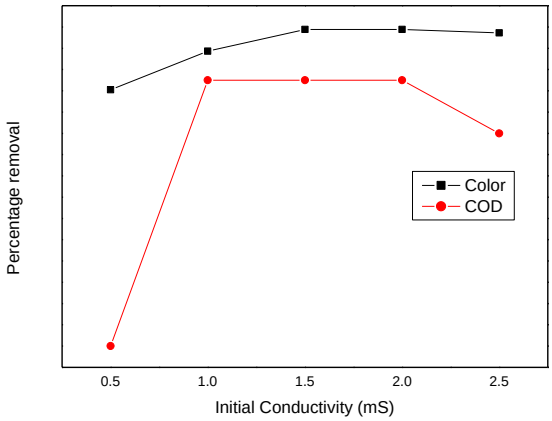

Fig. 3 Percentage removal of color and COD on varying initial conductivity

Optimization of Electrolysis Time on Color and COD removal

To examine the effect of operating time, the current density and pH will be kept constant and the time of electrolysis is varied from 10 min, 15 min, 20 min, 25 min and 30 min. The concentration of hydroxyl and metal ions generated on the electrodes affects the removal efficiency and the time of electrolysis decided the metal ions generation rate in

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the system from the electrodes. An increase in the electrolysis time leads to generation of more ions resulting in the higher COD removal efficiency of the wastewater *(Khandegar and Saroha, 2012)*. It has been observed from the experiments that there is a reduction in the removal efficiency of color and COD at too less and high electrolysis time. The reduction is because after optimum time excess of metal ions are contributed to the solution which results in the re-stabilization of metal ions. The optimum operational time for was 25 min. Fig 4 shows the percentage removal of color and COD by varying time of electrolysis.

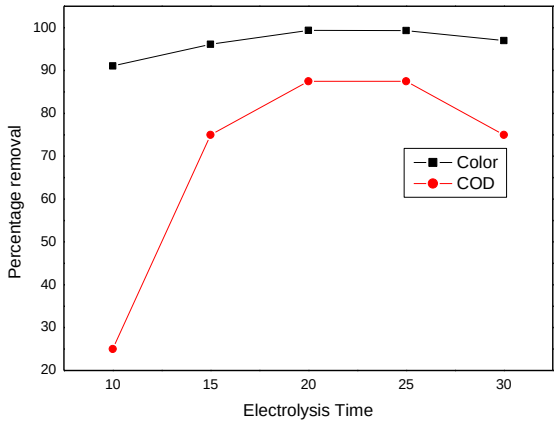

Fig. 4 Percentage removal of color and COD on varying electrolysis time

Effect of Initial pH on Color and COD removal

It has been observed that pH has not much significance in the color removal but higher COD removal efficiency was found near neutral pH for Malachite Green synthetic solution by electrocoagulation. Therefore optimum pH value was considered as 7 which is neutral. Fig. 5 shows the percentage removal of color and COD by varying initial pH of the solution. To study the effect of initial pH of the solution on the removal by the EC process, the pH was varied from 4 to 10. It has been observed that pH has not much significance in the color removal but higher COD removal efficiency was found near neutral pH for synthetic solution of dye by electrocoagulation. At pH 7, behavior was attributed amphoteric character of aluminum hydroxide which does not precipitate at very low pH. At higher pH efficiency reduces because at higher pH new aluminum complex forms $Al(OH)_4^-$ & $Fe(OH)_4^-$ which are soluble and poor coagulants and directly affects the pollutant removal *(Daneshvar et al., 2006)*.

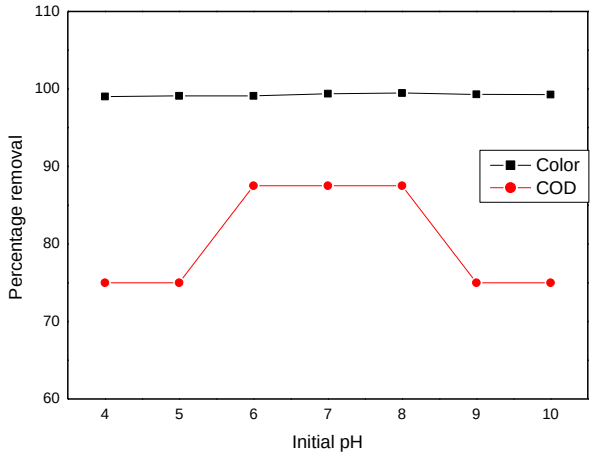

Fig. 5 Percentage removal of color and COD on varying initial pH

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Effect of Initial Dye Concentration on Color and COD removal

Concentration of dye is one of the important factors which decide the efficiency of the treatment unit. In the study, optimization of the operational parameter at usual concentration of dyes in textile industries i.e. 150 mg/l has been done. Under optimized operational conditions i.e. inter electrode distance (1 cm), initial conductivity (1mS), pH as 7 and Electrolysis time as 20 min, the concentration of dyes varied from 50 mg/l, 100 mg/l, 200 mg/l, 250 mg/l, and 300 mg/l. The effect of various concentration of dye on color and COD removal was investigated. It has been observed that there is not that much difference in the color removal from Malachite green dye and more than 99% color removal was achieved on varying concentration. In case of COD it is seen that lower the dye concentration higher the COD removal efficiency. This decrease in removal efficiency due to the formation of insufficient number of metal hydroxide complexes produced by the electrode to coagulate the greater number of dye molecules at higher concentrations *(Kobya, 2003)*. Therefore, it is quite clear that, the lower is the dye concentration the better is the decolorization efficiency. Fig 6 shows Percentage removal of color and COD on varying initial concentration of dye.

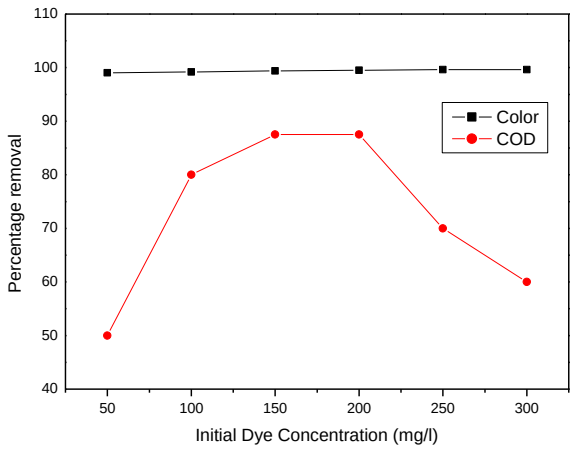

Fig. 6 Percentage removal of color and COD on varying initial dye concentration

CONCLUSION

In present study, an electrocoagulation lab scale set-up was constructed for the removal of Malachite Green dye from aqueous solution. Electrocoagulation using stainless steel electrodes for Malachite Green dye at optimum inter electrode distance of 1 cm, with initial conductivity and pH of solution as 1.5 m/S and 8 respectively for the electrolysis time of 20 min, 99.50 % and 85.71 % of color and COD removal was achieved respectively. These results showed that Electrocoagulation is efficient method for the treatment of the dye wastewater and the quality of the effluent was found to be better than the discharge limits according to the standards. This result confirms that the proper tuning of this method will be beneficiary for the removal of other dyes in large scales in industries.

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