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Removal of an Azo Dye Congo Red from Aqueous Solution by Electro Coagulation Associated with the In-Situ Adsorption Process

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Abstract **- Effective removal of Congo red from the aqueous solution has been studied using the electro coagulation technique using copper wire as both cathode and anode. As the voltage increases from 0.0 V to 55.0 V DC voltage, the removal efficiency increases up to 98% with the time limit of 35 seconds at a normal pH value of 7.0 - 7.85. As the temperature increases from room temperature to 75 ^{0}C, the coagulation increases and even the number of pairs of electrodes plays an important role in coagulation. The obtained sludge is characterized to identify its composition.**

Keywords: **Electro coagulation, PXRD, Congo Red, Copper Electrodes, Adsorption**

I. INTRODUCTION

After the industrial revolution, the world is facing many challenges due to industries. One of the major challenges is to provide clean and healthy water to the citizens or population throughout the world. The underdeveloped countries facing the most water problems due to some natural processes, human activities, and the effluent from industries [1]. A dye is also a source of water pollution. The Dyes have a wide scope of modern applications, for example, material coating, painting, calfskin printing, photography covering, beauty care products, pharmaceuticals, and photo-chemical enterprises. A dye is a characteristic or manufactured substance used to add shading or to change a shade of a substance. The industrial dyes are classified into anionic, cationic, and non-ionic dyes.

These coloring agents are carcinogenic, even sometimes mutagenic and direct releasing of dye contaminated wastewater from the textile, paper, pharmaceutical, and medical industries into freshwater bodies decreases the quality of water and even increases in the BOD and COD causing a bad impact on health also they are causing disease related to respiratory organs [2]. Typically, organic, synthetic, and physical techniques have been performed for color evacuation, such as chemical oxidation, ozonation, membrane filtration, biological treatment, coagulation, adsorption, and biosorption have been reported extensively, yet the natural procedures have not been extremely fruitful due to the non-biodegradable nature of general colors [3]. In coagulation acid, dispersive dyes, vat, etc can coagulate but they do not settle, cationic dyes were not coagulating during the coagulation. On the other hand, the chemical methods generate a considerable amount of sludge which itself needs further treatment [4].

One of the interesting methods of color removal is using physical adsorption. In the adsorption process, there must be adsorbate and adsorbent, the process occurs at the interface between the adsorbent and the contaminated water, but as the contact time increases after peak point desorption takes place. Most activated carbon is used in the adsorption method but the process of preparation is expensive and not easy to regenerate carbon [4], [5]. Some biological techniques are used to treat dye-contaminated water, but the dyes inhibit the growth of bacteria due to their power of avoiding penetration of light and other characteristics [6].

The drawbacks associated with the conventional techniques forced the industries/researchers for effective treatment methods for the complete removal of pollutants. As of late, there is an expansion in the number of color enterprises in this world and an increase in ecological danger. Many research papers have demonstrated the equivalent and discovered a few answers for this, yet some procedures have downsides like high upkeep, less proficiency, and even the cost matter for Small Scale Industries.

The electrochemical method involves electro coagulation (EC), electro Fenton, electro reduction, direct electro-oxidation [7]. EC is different from other methods due to the non-addition of coagulant externally and no secondary pollutants. Electro coagulation is an elective wastewater treatment that joins electrochemical procedures and multiple mechanisms with regular synthetic coagulation [8]. During the electrolysis of water fine bubbles of O and H are produced and move upwards and form a fine layer of bubbles around the electrodes. Reaction takes place between the bubbles and the molecules of dye, later they start settling at the bottom mechanically [9]. The sludge itself adsorb the dye molecule faster by destabilizing the colloidal dye molecule [10]. Even though chemical procedures have high effectiveness, they require some positive conditions to achieve that greatest point. Adsorption is likewise extraordinary compared to other innovations utilized in the color evaluation process, however, the removal of sludge is quite hard. EC depends on electrodes, the distance between electrodes, current density, and contact time [11]. As applied voltage and current density increase, the formation of these fresh flocs or sludge also increases and hence a higher amount of adsorption takes place because the sludge formed has a large surface area [12], [13].

Electrocoagulation is a standout amongst other processes that had been utilized nowadays, since it likewise having a few preferences like high efficiency, less upkeep, no such additives, requires less maintenance, safety, and amenability to automation, the required direct current can be obtained by conventional methods or using of solar cells [13].

In reality, this process is an alternative to the traditional coagulation system in which coagulant is generated in situ through the dissolution of a sacrificial anode [14], [15]. It's usually accepted that the EC manner involves six important successive stages: (i) metal ions start migrating towards oppositely charged electrodes (electrophoresis) and aggregate due to neutralization of charges. (ii) the cation or hydroxyl ion (OH^-) forms a precipitate with the pollutants; (iii) the cation interacts with OH^- to shape a hydroxide, which has excessive adsorption thus, forming bonds to the pollutant (bridge coagulation) (iv) the hydroxides form large lattice-like systems and sweep via the water (sweep coagulation); (v) pollutants are converted to much less poisonous species employing oxidation. (vi) dyes are eliminated with the aid of electro flotation or sedimentation and adhesion to hydrogen bubbles [16].

EC offers an option in contrast to the utilization of metal salts or polymers and poly-electrolyte expansion for breaking stable emulsions and suspensions [17]. Generally, it can be accomplished by killing the net charge of the colloidal particles by the expanded convergence of counter ions in the diffuse layer, instrument of charge balance, consequently diminishing the power with the generation of oxygen and hydrogen at cathode and anode [18], [19].

Removal efficiency is directly proportional to the amount of anode consumed. The freshly prepared $Cu(OH)_2$ has a larger surface area and can form a strong bonding. This sludge or flocs can be easily removed by filtration [20], [21]. During the removal of some organic compounds from contaminated water-containing toxins, toxic chlorinated organic compounds, and chlorides the sacrificial anodes have to be replaced periodically, but this procedure requires high electricity and results in a considerable amount of sludge production [10]. In most cases, the color and pollutant removal efficiency is about 95% and even it goes to 99%, to reach that point the solution must have good conductivity and this can be done by adding electrolyte [22].

Recent studies conducted by Vidya Vijay et.al [4] have concluded that 90% of removal is achieved with suitable salts, current density, and reaction time. Another team Ningning Liu et.al [7] concluded that electrolysis time plays a major role in removal efficiency and also reveals that aluminium electrodes are better than iron electrodes. Jorge Vidal and his co-workers [8] conclude that 75% of acid black-194 dye can be removed by applying high current density. The studies carried out by Ahangarnokolaei et.al [9] strongly conclude that the EC process did not depend on pH and the surrounding environment. Zazou *et al.,* [11] used iron electrodes and EC coupled with other processes to show that 100% of color removal efficiency along with turbidity and ToC (Total Organic Carbon) removal of 97% by EC-EF process. Ghalwa et.al [19] claims that high temperature is not favourable for coagulation.

In this present investigation, the electro coagulation process is adopted for the effective removal of Congo red from the aqueous solution using copper wire as electrode material. Copper electrodes are selected because of their high conductivity, least receptive, and modest expense. The voltage, electrolysis time, inter-electrode distance, temperature, and the number of pairs of electrodes have been studied and results are analyzed.

II. EXPERIMENTAL PROCEDURES

A. Materials and Instruments Used

The azo dye (Congo red) of Hangzhou Dimacolor Imp. & Exp. Co., Ltd. was taken in the form of powder. 60V DC regulated power supply TC-97 is used for DC voltage supply, insulated copper wire is purchased from the local market. The EC treated dye solution is centrifuged with a Centrifuge machine (Remi 24) for a fixed time of 10 minutes about 5000 RPM. The obtained centrifuged solution is analyzed using Uv-Spectrophotometer (Etico SL 159). The sludge generated during the electrolysis is characterized by utilizing XRD model Philips X-Ray diffractometer (PW/1050/70/76) with CuKα radiation (λ = 1.54Å) at room temperature and FTIR spectra are recorded using Brukers, FTIR-1500, in the range 400-4000 cm^{-1} using KBr as a pellet for calibration and opus 0.5 software for plotting.

B. Congo Red Dye and Preparation of 15ppm Dye Solution

Congo Red an azo dye C−N=N−C with Molar mass of 696.665 g/mol and the chemical formula is $C_{32}H_{22}N_6Na_2O_6S_2$ and an organic compound containing disodium salts of 3,3'-(1E,1'E)-biphenyl-4,4'-diylbis (diazene-2,1-diyl)bis (4-aminonaphthalene-1-sulfonate), used in the cotton textile industry to dye because they are resistant to washing and light, it is also used for staining in some of the biological processes for examine under a microscope and also used in tissue staining in histology.

Somehow it is also used as an acid-base indicator. Congo Red is an indicator dye that is blue-violet at pH 3.0 and red at pH 5.0. It is toxic to many organisms and is a suspected carcinogen and mutagen in some cases, at higher concentrations it metabolizes in the body to produce carcinogenic compounds such as benzidine. The structure of the Congo red dye is given in Fig. 1.

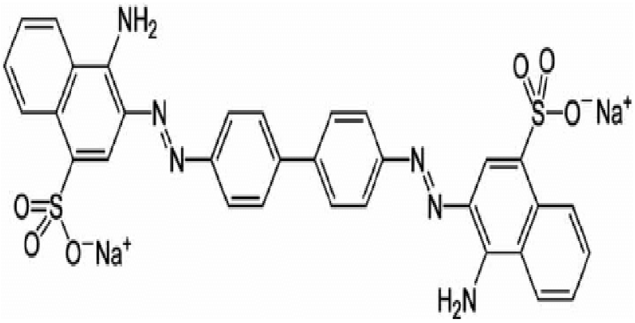

Fig. 1 Structure of Congo red dye

To investigate the efficiency of the electro coagulation process in dye removal, Congo red an azo dye is used as model dye. A 15ppm of a dye solution was prepared by taking 15 mg of CR dye and dissolved in 1000 ml of double-distilled water. The electro coagulation experimental setup is as shown in Fig. 2.

C. Preparation of Copper Electrodes and Coagulation Studies

The insulation or coating on the copper wire is removed and bent in the form of a coil of radius 0.5 cm, there are 8 turns obtained and the distance between each turn is about *0.2* cm. In the present study, the copper coil is used as both cathode and anode. To study the effect of the EC process, an experiment is carried out in the lab at normal room temperature between 25 ^{0}C to 28 ^{0}C. 150 ml of 15 ppm CR dye solution is taken in a plastic beaker of 250 ml, the electrodes were kept in the solution, then the DC voltage is applied across the electrodes which are separated by 0.5 cm. The electro coagulation is examined by studying the different parameters like initial dye concentration, voltage variation, contact time, number of pairs of electrodes, the distance between the electrode, and temperature. During the studies, after treatment the solution is taken in a tube and centrifuged with fixed time (10 minutes) and speed (5500 RPM), then centrifuged filtrate is taken in a dry test tube and dye removal efficiency is checked with Uv-Spectrophotometer (SL 159) and the removal efficiency is calculated using the equation

$$\text{Re}\,moval\ efficiency\ \ (\%) = \left[\frac{C_0 - C_e}{C_0}\right] \times 100 \quad \cdots(1)$$

Where C_O is the initial concentration and Ce is the equilibrium concentration.

The amount of anode dissolved (ϕ) in an electrolytic solution during the EC process can be calculated using Faraday's law

$$\phi = \frac{MIt}{nF} \quad \cdots(2)$$

Where I is the current through electrodes in mA, F is the faraday constant(96487Cmol^{-1}), n is the number of electrons, M is the molecular weight of the metal used, t is the saturation time, and by knowing these things ϕ can be calculated.

Energy consumed during the electrolysis is given by:

$$E = U \times I \times t \quad \cdots(3)$$

U is the applied voltage (V), I be the current expressed in mA, t is electrolysis time.

Pollutant removal was represented by adsorption phenomena and the amount of pollutant adsorbed by

$$q_e = \frac{(C_0 - Ce)V}{M} \quad \cdots(4)$$

where V represents volume dye solution, Ce and C_0 stand for equilibrium and initial concentration of dye solution (mg/L) respectively, M stands for the quantity of anode dissolved (g).

Specific Energy Consumption (SEC) can be calculated using.

$$SEC = \frac{U \times I \times t}{V(C_0 - C_e)} \quad \cdots(5)$$

Where V is the volume of a dye solution, C_o and C_e are the initial and final concentrations.

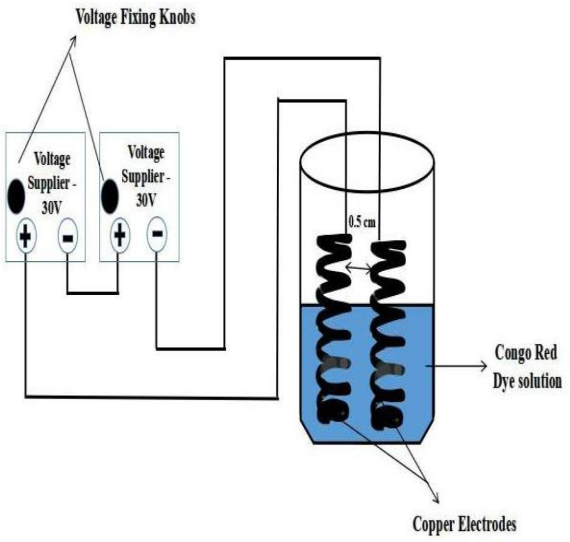

Fig. 2 Schematic Electro Coagulation Experimental Setup

The reactions that take place during electrolysis is given by

Anode: $Cu \rightarrow Cu^{2+} + 2e^-$ …..(6)

Cathode: $2H_2O + 2e^- \rightarrow H_{2(g)} + 2\,OH^-_{(aqua)}$ …..(7)

Formation of Metal Hydroxide

$$Cu^{2+} + 2OH^{-} \rightarrow Cu(OH)_{2(s)}.\ldots\ldots(8)$$

III. RESULTS AND DISCUSSION

A. Effect of Voltage Variation

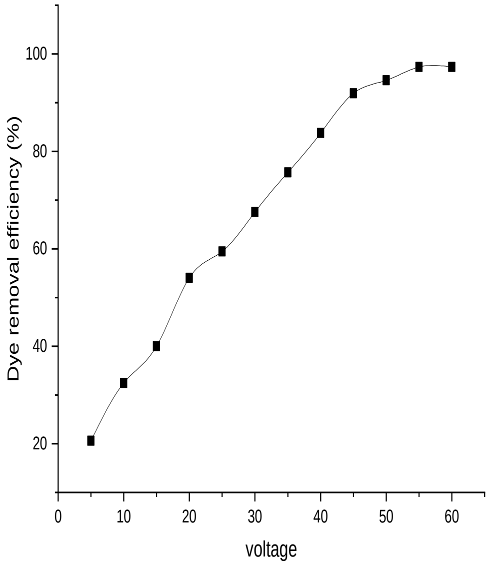

Fig. 3 Effect of Voltage on Coagulation

Fig. 3 shows the variation in the removal efficiency of dye with voltage. The 150 ml of CR dye solution is taken in a beaker of 250 ml. The voltage is applied across electrodes, later the solution was taken in a tube and it is centrifuged for a fixed time (10 minutes) and fixed rotation (5500 RPM) and readings were taken using Uv-Spectrophotometer (SL 159) and it is found that dye gets coagulated/adsorbed on a sludge. As the DC voltage varies from 0-60V DC the removal efficiency increases.

As the voltage increases, more metal ions are formed and they recombine with hydroxyl ions to form more metal hydroxide which adsorbs the dye molecules. It is also found that, with an increase in voltage the bubble formation also increased, further this indicates more hydrogen gas liberation [14]. The energy consumption increases from 2.22 kWh to 264 kWh along with electrode consumption from 0.22 mg to 18.0 mg with an increase in voltage. Specific energy consumption will also increase from 0.036 kWh (SEC for 5V) to 4.63kW/h (SEC for 55V). At 0V there is no removal and from 55V onwards there is 98.89% removal of the dye from the contaminated water. It is noted that for 55 and 60V dc voltage showed the same removal efficiency hence it may conclude as a saturated voltage range for efficient dye removal. So, 55V is taken as the optimized voltage for further work.

B. Study of Electrolysis Operating Time

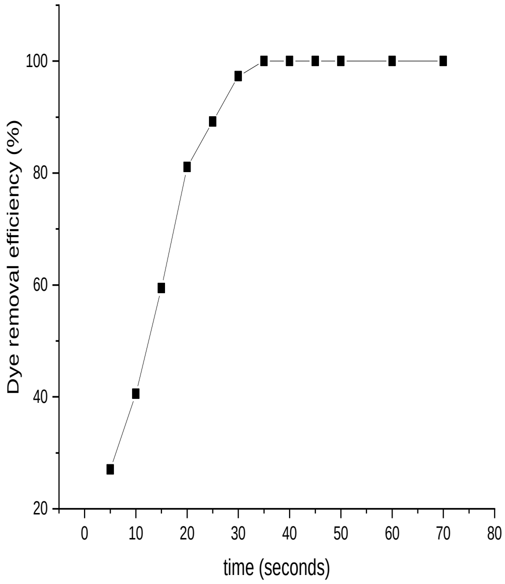

Fig. 4 Effect of Operating Time on Coagulation

In the present study, the operating time for electro coagulation is studied from 0-70 seconds by applying the optimized voltage 55V in a 15 ppm dye solution of volume 150 ml. For each electrolysis time, the dye removal efficiency is calculated and it is shown in Fig.4. It is found that in dye solution, most of the dye gets coagulated at 35 seconds. As the time increases above 35 seconds, the removal efficiency is almost constant. This might be due to effective adsorption of almost all the dye molecules.

In the beginning, the formation of copper hydroxide (sludge) is less and hence the decolorization rate is less. As electrolysis time increases more current passes through the electrodes, and the discharge of metal ions also increases which is converted to metal hydroxide in the solution so that sludge generation is high as the time increases and hence the removal efficiency increases [15], [16].

It is noted that the froth or the flocs become massive and settle down, leaving the colorless solution without a centrifuge. The Energy consumption also rises from 330Wh at 5 seconds to 3.9 kWh at 60 seconds but it consumes 2.3 kWh for 35 seconds. The anode consumption also rises from 1.97 mg to 24.0 mg as the time increases from 5 seconds to 60 seconds. At 35 seconds of electrolysis time, anode consumption is 13.83 mg and these characteristics are shown in Table I.

C. Effect of Change in Initial Dye Concentration

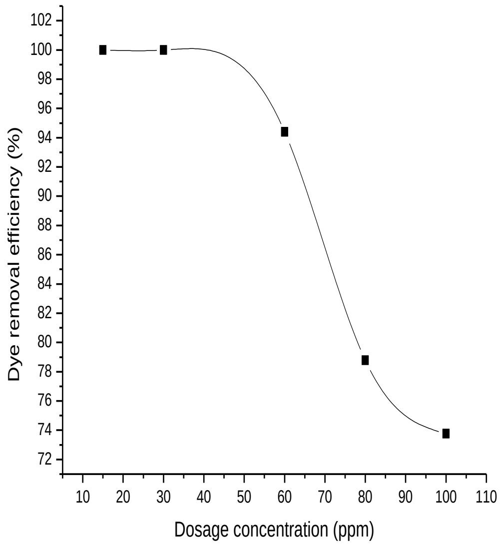

Fig. 5 Effect of Change in dye Concentration

It is evident from Fig. 5 that as the dye solution concentration increases from 15 ppm to 100 ppm at optimized conditions, the removal efficiency decreases from 98% (for 15 ppm) to 73% (for 100 ppm). The decrease in efficiency because of the presence of more dye molecule in the electrolytic solution, so that the number of ions released from the electrodes which form metal hydroxide (sludge) not sufficient to coagulate the 100 ppm dye solution within 35 seconds and also the formation of the in-situ adsorbent by electrodes is not sufficient to absorb the dye molecule, but it can be removed with an increase in time so that sufficient adsorbent is formed inside the solution [17-19].

D. The Efficiency with the Number of Pairs of Electrodes

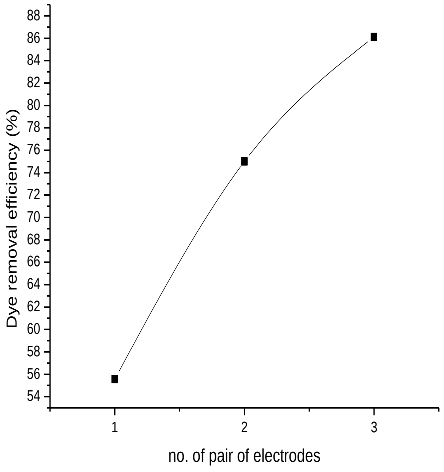

Fig. 6 Effect of the Number of Pairs of Electrodes

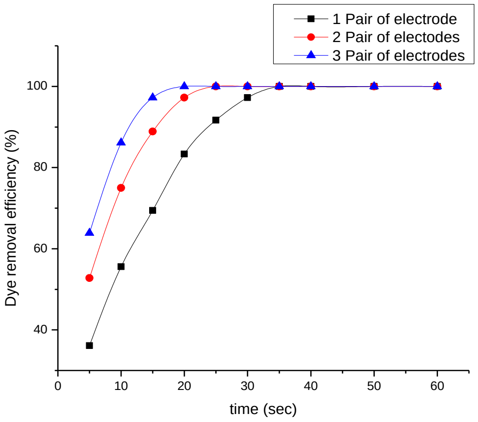

Fig. 7 Removal Efficiency is based on a Number of Pairs of Electrodes

Fig. 6 shows that, as the number of pairs of electrodes increases from 1 to 3 the removal efficiency increases at an electrolysis time of 10 seconds. This may be due to the availability of more and uniform distribution of copper hydroxide all over the electrolytic solution at 55V DC. The variation of dye removal efficiency with time at different numbers of pairs of electrodes is investigated and results are indicated in Fig. 7. This signifies that, as the number of electrodes increases the removal efficiency also increases and the electrolysis time is reduced. When more than one pair of electrodes are introduced into the solution the total area covered is quite more and the voltage is almost the same throughout the solution compared to a single set and hence influences the number of ions around them. On the other hand the formation of sludge also becomes high because of the more sacrificial anodes, in-situ production of more copper hydroxide adsorbent from these electrodes helps in fast coagulation. If further the pair of electrodes increases, the production of adsorbent also increases. The electrode dissolution also increases because of more electrodes, but the SEC and energy consumption are 2.70 kW/h and 154 J respectively.

E. Effect of Temperature on the Coagulation Process

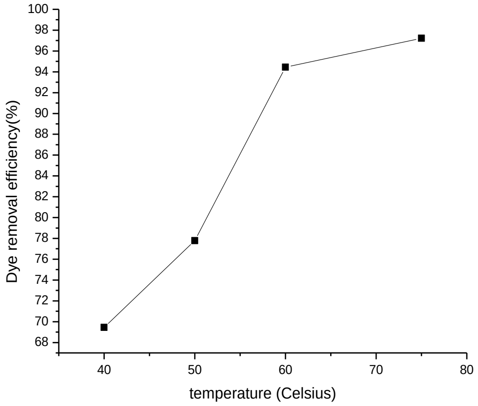

Fig. 8 Variation of Color Removal Efficiency due to Temperature

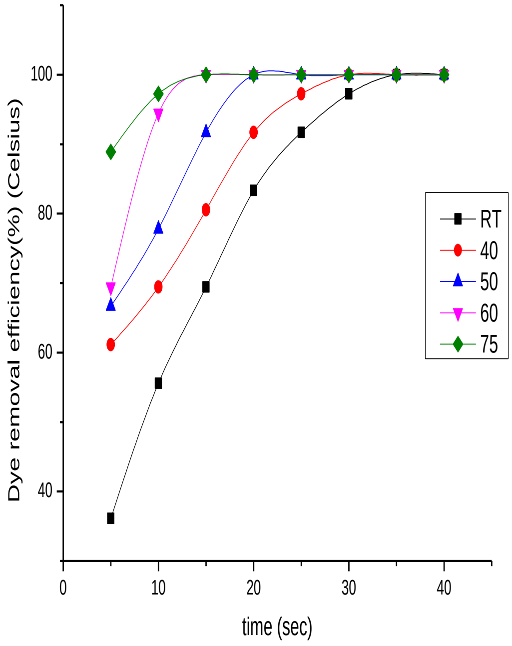

Fig. 9 Effect of Temperature on Coagulation

Temperature plays an important role in the removal of CR dye using the electro coagulation process. Fig.8. represents the variation of dye removal efficiency with respect to temperature at 15 seconds of electrolysis time. As the temperature rises from 40 ^{0}C to 75 ^{0}C, the removal efficiency rises from 69 % to 98 %. This is because as the temperature of the solution increases, the kinetic energy of each molecule increases and they tend to move faster because of kinetic energy [16 - 20].

At a particular temperature, 55V DC is introduced into the solution. Due to increased temperature the electrodes itself gets heated and at the same time the ions released from the electrodes also move faster in different directions, so that the momentum and distribution of the ions are quite larger compared to normal room temperature and hence more collision between the dye particles which could easily form a precipitate and settle down.

It is evident that an observable change in the temperature of the solution or its surroundings greatly influences the coagulation and removal efficiency [20]. Whereas some literature surveys raised that, removal efficiency decreases with an increase in temperature and claimed that the adsorption process is exothermic [15-19].

Fig. 9 Shows the variation of dye removal efficiency with time at that particular temperature and it signifies that as the temperature increases removal efficiency is higher and the required electrolysis time also reduces.

F. Role of Electrode Distance on the Removal Efficiency

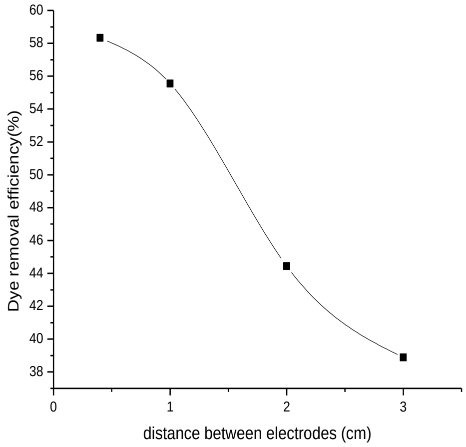

Fig. 10 Effect of Electrode Distance

Fig. 10 gives evidence that as the distance between the electrodes increases, the efficiency decreases. At 0.5 cm electrode distance, the removal efficiency is 59% and when they are placed 3 cm apart, the removal efficiency decreases to 39% at optimized voltage and electrolysis time. The decrease in the efficiency is because of the increase in resistance (number of bubbles formation is very less) between the electrodes i.e. when they are placed apart at long distance, immigration of ions between the cathode and anode is very less and hence there is a decrease in the voltage in the solution so that the formation of the sludge also decreases with increase in distance and hence amount of sludge also decreases but power consumption is high. When the distance between the electrodes is small the resistance is small (More bubbles with large size) and voltage across the solution nearly equal to the applied voltage and hence results in flocculation. The energy consumption is less when they are placed close to each other compared to the energy consumed when the electrodes are kept at 3cm apart [16 - 21]. But the aging of the equipment is faster. When the electrodes are kept very close to each other they may get shorted and the result is damaging to the whole system Table I gives the characteristics of the EC method at optimum conditions such as 55. 0 voltage and electrolysis time of 35 seconds and the results are mentioned in Table I.

TABLE I CHARACTERISTICS OF THE ELECTROCOAGULATION METHOD AT OPTIMUM CONDITION

EC	**Value**
Anode dissolved (ϕ)	13.83 mg
Energy consumption (E)	2.31 KWh
Current density	1.2A/cm^2
Specific Energy Consumption (SEC)	2.66Kwh
Amount of pollutant adsorbed (qe)	18.46 mg

IV. CHARACTERIZATION OF SLUDGE

A. Powder X-Ray Diffraction Pattern of Sludge

To identify the chemical composition, PXRD spectrum of the sludge is obtained. After the treatment of water, the sludge is separated using filter paper and heated in a furnace at 90^{0}-150^{0}C for 5 minutes and the XRD spectrum is taken (Fig.11). It is found that, the presence of CuO, Cu_2O, and $Cu(\breve{O}H)_2$.

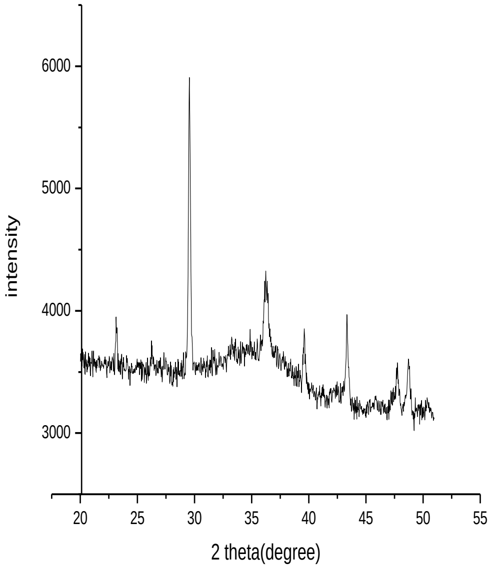

Fig. 11 PXRD pattern of Adsorbent

The peak at 23^{0} shows that there is a presence of $Cu(OH)_2$ [JCPDS file no. 35-0505] which is an Insitu adsorbent that absorbs dye from the synthetic dye solution. There is also a presence of CuO at 36^{0}, 40^{0}, $44^{0,}$ and 48^{0} [JCPDS file no. 01-080-1916] and the main peak at 29^{0} is Cu_2O [JCPDS file no. 05-0667] because the copper hydroxide is metastable so that it converts to copper oxide. No other product formation is observed.

B. FTIR Spectroscopy

To find out whether the $Cu(OH)_2$ is formed or not and dye is adsorbed on it, the resultant sludge is characterized. The FTIR spectrum of sludge is shown in Fig.12. The band at 3356.67 cm^{-1} gives $Cu(OH)_2$ and also can be assigned to various functional groups like (N-H), (O-H), and (NH_2). The other bands at 712 cm^{-1} and 491cm^{-1} show the presence of CuO, this is because the sludge is heated around 90^{0}-150^{0}C. The peaks at 502 cm^{-1} give the spectra of $Cu(OH)_2$. The peaks at 712 cm^{-1}, 461 cm^{-1}, 2924 cm^{-1} and 1043 cm^{-1} indicate the presence of Congo Dye [23], [24]. The CU_2O peaks are observed at near 1409 cm^{-1}, 1051 cm^{-1}, 3423 cm^{-1}, 2917 cm^{-1} and at 1399cm^{-1}.

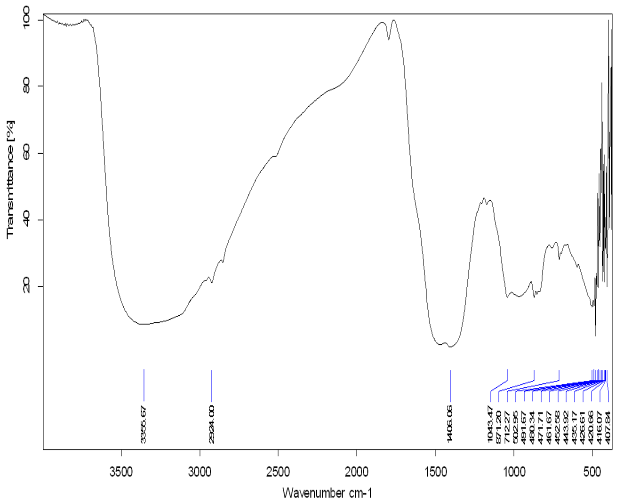

Fig. 12 FTIR spectrum of coagulated sludge (adsorbent)

V. CONCLUSION

After going through the studies, it is found that the potential of copper electrodes in the process of dye removal is sufficient to remove 98 % dye from the synthetic dye solution and it is verified by various parameters like operating time, initial concentration of the dye, voltage, temperature, number of pairs of electrodes and the distance between the electrodes. The metallic and hydroxyl ions from the electrodes react and form metal hydroxide which promotes faster adsorption because of having a larger surface area. This method has several advantages over the other process in terms of lower desorption or no desorption rate, efficiency, and the amount of required adsorbent and even it has some demerits. In large-scale industries, this method can be adopted at a cheap cost. At any time above 60 seconds of saturation, the sludge can be removed physically without centrifuge as it easily settles down.

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