# Non-thermal plasma as a new advanced technique for intensifying the industrial wastewater treatment

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### **ABSTRACT**

This manuscript investigates an opportunity of improving the dye elimination from wastewater by non-thermal plasma. Computer simulation technology (CST) was utilized for the double pin-to-plate configuration in 3D software simulation package for prediction field and voltage among the high voltage and the ground electrodes. The outcomes indicated that optimal gap separation for non-thermal plasma is about 5 mm. This optimal distance was used for corona region and ozone production. Experimental arrangement of double pin-to-plate discharge method has been deliberate examined the elimination of Acid Blue 80 dye. The influence of contact time, initial concentration, solution pH and gap distance have been established on Acid blue removal efficiency. The color removal performance using corona discharge displayed 80% dye color elimination competence within 10 min. The whole color removal was attained after 35 min for diverse concentrations from 5–100 ppm. In addition, the corona current performance at different initial solution pH was presented. Furthermore, the discharge energy and corona current were measured at different gap distances. Energy yield and electrical energy per order (EE/O) for decolorization in diverse gap distance were considered.

*Keywords:* Corona discharge; Ozone generation; Non-thermal plasma; Wastewater treatment; Dye removal

#### **1. Introduction**

There are serious difficulties through ecological contamination of water created by textile productions in the world. In old-style treatment methods textile polluted water were treated by either chemical or biological methods. Conversely, these techniques have time and price difficulties for water treatment regarding to the requirement of big facilities and existence of some pollutants

that are not simply degradation. Therefore, the evolution of advanced water-treatment tools that can resolve these difficulties is vital. The best solution for dye contaminated water is the ozone treatment [1]. Artificial dyes are usually utilized as a wellspring of shading specialists in material, cowhide, coloring, makeup, paper and nourishment handling businesses because of their imperviousness to biodegradation and shading adaptability. Azo dyes, described that type has the azo assembly and comprising of two nitrogen atoms as the chromophore in their atomic structure. Azo dyes have propelled warm

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and optical properties because of their concoction nature, sub-atomic size, and structure. The release of color containing effluents into water bulks may be cause genuine ecological and well being issues. The effluents are poisonous and cancer-causing with a mind boggling structure fundamentally in light of aromatic amines that created by-results of their incomplete degradation [2]. Recently, the applications of plasma-predicated oxidation methods on the degradation of pollutants have been increasingly explored owing to their involution, diversity and high oxidant capacity. Diverse strategies (e.g., coagulation, ion exchange, electro coagulation and flocculation, layer partition, biosorption, bioaccumulation and oxidation) have been utilized with shifting degrees of achievement for the removal of colors and different contaminants from water and wastewater. Real weaknesses of these techniques, particularly for color expulsion, incorporate halfway corruption, high cost and particular confinements. Accordingly, there is a basic need to create strategies to productively treat color containing wastewaters before releasing them into the environment. As of late, propelled oxidation forms (AOPs) have drawn in interest as productive strategies for removing natural colors from wastewater. The primary system of cutting edge oxidation procedures is an in situ time of extremely solid oxidizing operators (e.g.,  $OH^{\circ}$ ,  $O_{3'}H^{\circ}$ ,  $O^{\circ}$ ,  $O_{2'}H_2O_2$  and H2 ) and additionally synthetic and physical procedures that furnish consequent response with the toxins, bringing about the decay of broke up atoms in water through oxidation responses [3,4]. Non-thermal plasma corona discharge for wastewater organic mixtures reports the next compensations: more preponderant competence, extra charge effective tools, and more facile process. Furthermore, no other contaminants are produced since the innovation takes after the green science rules in the entire corruption process. The main target of this article is to observe the capability of non-thermal corona discharge plasma to generate ozone and active radicals for treatment the wastewater polluted with Acid Blue dye pollutant. Investigational were supported out to define the impact of gap separation, contact time, pH, initial concentration on dye removal. Theoretical framework for the generation of non-thermal corona discharge plasma was researched utilizing 3D programthat include the integral equations technique (IET)[5,6] to resolve the partia1 derived function equivalence that define the performance of electrostatic fields. Furthermore, the discharge energy and corona current were measured at different gap distance. Energy yield and electrical energy per order (EE/O) for the decolorization process in diverse gap distance were considered.

### **2. Materials and methods**

### *2.1. Corona reactor*

The double pin-to-plate configuration was fabricated from 2 mm diameter of the pin with 4 cm length and the ground conductor is rectangular with dimensions 50\*60\*0.2 mm and total area 30 cm<sup>2</sup>. High-voltage of 15 kV was used in two pin electrodes, while the plate was grounded as seen in Fig. 1 [7,8].

Pulsed high voltage utilized as a part of this article comprise from PWM circuit based on the 555 timer and ignition curl FTM063GT-GC5MIC2000. PWM circuit appeared in Fig. 2 with the electronic drawn for the circuit utilized. The operation work of the PWM circuit begins with the circuit powers up the oscillator cycle then the capacitor C1 to charge through the right half of R1 and diode D2. At the point when the voltage on C1 achieves 2/3 of +V, the limit is initiated, which thus causes the yield, and release the capacitor C1. At the point when the capacitor C1 begins to release through the left half of R1 and D1 the voltage on C1 falls beneath  $1/3$  of  $+$  V, the cycle repeats. Capacitor C1 charges through one side of  $R_1$  and releases through the other side. The aggregate of the charge and release resistance is dependably the same; along these lines the wavelength of the yield sign is consistent. Just the obligation cycle fluctuates with  $R_1$ . The general recurrence of the PWM signal in this circuit is dictated by the estimations of  $R_1$  and  $C_1$ . The IRFZ46N MOSFET work relies upon the 555 clock signal, when the MOSFET work utilized for isolated the DC source and nourish the ignition loop. Ignition loops are utilized as a part of autos to create a flash which starts the burning of the fuel-air blend in the chambers.

An ignition coils; similar to transformer involves an iron center with an essential and an auxiliary winding. The turn's proportion amongst auxiliary and essential is in the request of 100:1. Two windings are associated toward one side, so that the auxiliary is naturally grounded. The most extreme voltage contrast in this manner is about 60 kV, which is sufficient to hop around 10 cm air hole. Arrangement of double high-voltage electrodes has been planned and examined experimentally for its consequence on the decolorization of Acid Blue. High voltage of 15 kV peak output was applied to pin electrode through frequency 100 Hz, the period time 10 ms, the rise time equal 70.46 µs, positive width 8.615 ms and negative width 1.376 ms.The wave form of the switch signal feed IRFZ46N MOSFET, the output voltage and corona current are exposed in Fig. 3.

The double electrode of high voltage separated by 5 mm gap recorded from the simulation work is the best distance



Fig. 1. Corona reactor system.



Fig. 2. PWM Circuit Based on the 555 Timer.



Fig. 3. Output voltage, switch signal and corona current.



from the ground plate. Fig. 4 shows the non-thermal corona discharge plasma region. HITACHI U-3900UV spectrophotometer has been used to measure absorption of the dye before and after experimental at wavelength of 640 nm [9,10]. Decolorization capability is determine as the relation of the removed absorption to the initial absorption:

Decoloration competence

$$
= \left(\frac{\text{Initial absorption} - \text{Absorption after treatment time t}}{\text{Initial absorption}}\right) * 100 \quad (1)
$$

# *2.2. Chemicals*

Acid blue 80 dye was prepared by liquefying the suitable quantity of the dye in purified water. Molecular formula is  $C_{32}H_{28}N_2Na_2O_8S_2$  with 678.68 g mol<sup>-1</sup> molar mass and its chemical construction is shown in Fig. 5. Acid blue 80 is consumed for, polyamide fiber, fleece, silk and its mixed fabric coloring, Fig. 4. Spark discharge from pin electrode. tops, coloring scattered hair, yarn bundles, socks and weav-



Fig. 5. Molecular structure of Acid Blue 80 Dye.

ing yarn, additionally it can also be used in leather dyeing. The dye solution pH was adjusted at various initial values using either sodium hydroxide or sulfuric acid solutions.

#### **3. Simulation model**

Non-thermal corona plasma in air motivated toward a little zone in the area of the little radius wire. Proposed model using 3D software package was utilized to discover the optimal gap distance with 15 kV. The field and voltage appropriation in the gap are defined, in order to determine the mechanisms of corona release and ozone produced. To understand the mechanisms of corona discharge and generation of ozone, active ridicules should be known and the field and voltage in the gap space between the two electrodes should be determined. The best way to optimize the corona devices is to known the onset voltage and onset electric field that can determine by the voltage and field [11–14]. The field has important influence on the producing of corona discharge. To produce the sequence reaction the field should be strong enough that producing the electron from all the type of collision (ionization, photoionization, electron emission). Also, the strong field accelerates the particles motion between two electrodes and prevents the recombination between ions and electrons. So, the 3D programming that settle the Maxwell's conditions was used by utilizing the finite integration technique to disclosure out the ideal cathode crevice for a given state of 15 kV. The deliberate voltage source appeared in Fig. 6 has been utilized to nourish the proposed framework showed in Fig. 7.

The effect of fluctuation the gaps from 1 to 20 mm with step 1 mm on the maximum field closed to the high voltage electrodes has been investigated in Table 1 and Fig. 8.

It has been noticed from Fig. 8 that the space of 5 mm satisfies maximum field. At each studied decolorization time, the location of the maximum field for each distance is presence near to high voltage electrode. It's noticed that the shape of field around the sharp pins with high values compared with all framework, so this area expected



Fig. 6. Voltage waveform feed the software programs.



Fig. 7. Simulation model.

high corona. As displayed in Fig. 9 the field was present at different lines above the water surface along the diameter of the corona reactor. It is noticed from Fig. 9 shape of field around the high voltage electrode contrasted with all items in framework, so local high corona area was expected. Shaded plot for the voltage, field, energy distribution and current distribution are presented in Figs. 10–13 respectively.

#### **4. Dye decolorization process**

The non-thermal plasma corona discharge process has great quantity of diverse active species and radicals that improve its efficiency. Release of these radicals in water deliver the rich sources of the various oxygen (ROS) and nitrogen (RNS) chemically active species, hydroxyl radical (OH° ) and hydrogen particles (H) from water atoms. In moist air, non-thermal plasma corona discharge produces







Fig. 8. Voltage waveform feed the software programs.

ozone  $(O_3)$ , hydrogen peroxide  $(H_2O_2)$ , singlet oxygen  $(O)$ , hydroperoxyl radical  $(HO_2)$ , superoxide  $(O_2^-)$  ions, hydrogen peroxonitrate (ONOOH), NO and NO<sub>2</sub>. It was exposed that the principle reactive species elaborate in dilapidation of organic pollutions are OH $^{\circ}$  radical and H<sub>2</sub>O<sub>2</sub> [15,16]. Especially, the OH° radical is distinguished to assume an essential part in debasing natural mixes since its oxidation potential is greater than that of nuclear oxygen and ozone.







Fig. 10. Voltage distribution.



Fig. 11. Field distribution.



Fig. 12. Energy density distribution.



Fig. 13. Current density distribution.

It was presumed that the  $NO/NO<sub>2</sub>$  molecules were produced in the discharge zone as a result of the reaction of nitrogen coming from the atmosphere with oxygen. The point by point creation procedures for these sorts amid the release are as follows [17–19]:

$$
e^- + H_2O \to OH^{\circ} + H^{\circ} + e^-
$$
 (2)

$$
e^- + O_2 \rightarrow O^{\circ} + O^{\circ} + e^-
$$
 (3)

 $e^- + O_2 + M \rightarrow O_2^- + M$  (4)

$$
\varepsilon^{-} + O_2 \rightarrow O_2 + 2e^{-}
$$
 (5)

 $H_2O_2^+$  hv  $\rightarrow$  2OH<sup>°</sup> (6)

$$
\mathrm{O}_3 + \mathrm{H}_2\mathrm{O}_2 \longrightarrow \mathrm{OH}^\circ + \mathrm{O}_2 + \mathrm{HO}_2^\circ \tag{7}
$$

$$
2H_2O + e^- \to H_2O_2 + H_2 + e^-
$$
 (8)

$$
O^{\circ} + H_2O \to OH^{\circ} + OH^{\circ} \tag{9}
$$

$$
\mathrm{OH}^\circ + \mathrm{H}_2\mathrm{O}_2 \longrightarrow \mathrm{H}_2\mathrm{O} + \mathrm{HO}_2^\circ \tag{10}
$$

$$
O_3^{\dagger}HO_2^{\circ} \rightarrow OH^{\circ} + O_2 + O_2^{-}
$$
 (11)

$$
O_3 + hv + H_2O \to H_2O_2 + O_2 \tag{12}
$$

$$
O^{\circ} + O_2 + M \to O_3 + M \tag{13}
$$

$$
N_2 + O = NO + N \tag{14}
$$

$$
N + O_2 = NO + O \tag{15}
$$

 $NO + O + M = NO<sub>2</sub> + M$  (16)

$$
NO + OH = HNO2
$$
 (17)

$$
NO2 + OH = HNO3
$$
 (18)

$$
2NO_2 + 3H_2O = NO_3^- + NO_2 + 2H_3O^+
$$
 (19)

$$
NO2- + H2O2 + H3O+ = ONOOH + 2H2O
$$
 (20)

Experiments were approved out to define the consequence of diverse features: gap separation, contact time, pH, and initial concentration on colour elimination capability of the dye. Fig. 14 displays the appearance of the acid blue dye solutions earlier and later the treatment process.

# *4.1. Effect of gap distance*

Separation among two conductor electrodes accepts a key part on plasma discharge treatment technique. Table 1 exhibits the most compelling field for various partitions by computer simulation technology (CST) 3D programs the results showed that gap of 5 mm gives the best field at the sharp tip of pin terminal. In order to check the effect of gap separation on the plasma discharge process, 1, 2, 5, 10, 15, and 20 mm gap distance are tested for volume of solution 100 ml (10 ppm), operation voltage15 kV, pH 6 at room temperature and pressure. Fig. 15 exhibits that 25 min represents the ideal time for the decolorization procedure. It appears from this figure that the expansion at the detachment enhances the shading decolorization process until 5 mm with further augmentation out there the shading expulsion efficiency decreased. The relation between the current flow and the color removal efficiency at diverse gap distance was investigated in Fig. 16. It was evident that increases the current flow in the circuit will be reflect on decreasing of the acid blue dye color removal. Also, vice versa is correct, with reducing the current flow in the circuit the acid blue dye color removal will increase. This result is in accordance with the expected results because the reduction of the current diminishes the amount of charge per time that flow in the ground plate. Such situation gives more chance to create the charge in the solution that may lead to more collision inside the



Fig. 14. Acid Blue earlier and later the treatment.



Fig. 15. Gap distance on color removal competence of Acid Blue dye.



Fig. 16. Relation of decolorization efficiency with corona current at different of gap distance.

solution and to generate more actives radicals that used in degradation of the organic dye. By utilizing the digital storage oscilloscope (Tektronix TDS2014), current probe (Tektronix A6021) and high voltage probe (Tektronix P6015A), the current was measured at various crevice separation. It can be reasoned that 5 mm is the best air hole detachment that confirmed most extreme proportion on shading evacuation fitness of 80% decolorization within 10 min. The aggregated dye removal was achieved completely within 35 min. Fig. 16 demonstrates that these outcomes might be added to certainty that as in-wrinkling in crevice removes the force of non-thermal plasma corona discharge diminish, which influence the era of artificially dynamic species, for example,  $H^{\circ}$ , O°, OH $^{\circ}$  radicals and hydrogen peroxide  $H_2O_2$  atoms. The artificial dynamic sorts could be used for the end of lowlevel natural contaminations.

### *4.2. The energy yield electrical energy per order (EE/O)*

The energy yield is a basic element for assessing the treatment procedure. It is the active radicals produced per 100 eV from electrical vitality include the procedure. It is of water surface under the pulsed corona discharge plasma system. It was calculated by using the following equation [20,21]:

$$
G = \frac{C_0 * V_0 * D\%}{100 * P * t}
$$
\n(21)

where G is energy yield (g  $kWh^{-1}$ ); P – power of the reactor (w),  $V_0$  – volume of treated solution (L),  $V_0 = 100$  ml, t – time required (s),  $C_0$ – concentration of the pollutant at t  $= 0$ , C<sub>0</sub> = 10 ppm.

The energy yield computed utilizing Eq. (14), where the power of the reactor equal root main square of the input current and voltage in the corona reactor. Fig.17 gives the joining of the energy yield  $(g kWh^{-1})$  and the time under various gap distance of acid blue dye arrangement. It is seen that the energy yield of the gap space 5 mm represents the highest energy yield compared with other gap distances.

Feature assessment of electrically behavior determined the treatment procedures are the electrical energy per order  $(EE/O)$  or the energy yields  $(g kW<sup>-1</sup>)$  of significance abatement are regularly utilized for some compound debasement or oxidation forms. It was indicated that the electrical energy per order given a straight association with the electrical capability (slight qualities mean additional compelling procedure) of an advanced oxidation process (AOP) framework, autonomous of the framework. EE/O was distinct as the electrical energy in kWh expected to prompt the corruption or oxidation of dirtied C by one request of extent in 1 m3 (1000 L) of contaminated water or air. EE/O principles (in  $kWh$  m<sup>-3</sup>) can be fined from the following Sunga relation [20]:

$$
E_{E/o} = \frac{P * t * 1000}{60 * V * \log(\frac{C_i}{C_f})}
$$
(22)

where *T* is treatment time *t* (min),  $C_f$  – final concentrations (mol  $l^{-1}$ ), *P* – power (kW), *V* – volume of water (L),  $V_0 =$ 100 ml;  $C_i$  – initial concentrations (mol l<sup>-1</sup>),  $C_i$  = 10 ppm.

The electrical energy per order was calculated from Eq. (15). Fig. 18 shows the relation between the electrical energy per order and the time under diverse gap distances of acid blue solution.



Fig. 17. Relation of energy yields with time at different gap distances.



Fig. 18. Relation between electrical energy per order with time at dif-

It was indicated that the EE/O expanding with expanding gap distance from 1 mm to 20 mm at the equivalent treatment time. The EE/O for gap distance 5 mm represents the lowest one compared with other gap distances. EE/O is most noteworthy utilized for conditions where early fixation is little (i.e., cases that are general first-arrange in focus) subsequent to the amount of vitality expected to transport around a reduction by one request of greatness by Lukes [20] and Sunga [22].

#### *4.3. Effect of contact time*

Impact of contact time is vital factor that impacts the response time among the pulsed discharge and acid blue color. This factor is recognized utilizing ideal foreordained separation. Fig. 19 demonstrates the consequences of the rate of color removal and contact time over studied time intervals of 0–35 min. Where, the volume of the solution 100 ml, operation voltage15 kV, pH 6, room temperature and pressure at initial concentrations 20, 40, 60, 80 and 100 ppm. The amount of dye decolorization is great at the initial periods for all studied dye concentrations. The initial 10 min recorded around 80% dye decolorization for all studied concentrations. As the contact time was increased, the dye decolorization was incremented until completeness within 35 min. It can be clarified plainly that the decolorization productivity of color increments through the expanding of contact time among the color and crown release created in plasma release framework, it achieved balance at 35 min.

## *4.4. Effect of dye concentration*

The most essential factor impact on the productivity of the release plasma treatment procedure is the concentration of acid blue dye solution. Analysis is performed by variation the initial dye concentration from 5 ppm to 100 ppm using volume of the solution100 ml, operation voltage15 kV, pH 6, room temperature and pressure. The quantity of color removal was reduced with increasing the initial dye concentration as appeared in Fig. 20. This may be cleared up by the way that color particles tend to take up through expanding shading center to gatherings of low diffusivity, this conveys down the measure of shading scattering to the anode



Fig. 19. Influence of contact time on color removal of Acid Blue dye.



Fig. 20. Influence of concentration on color elimination efficiency of Acid Blue dye.



Fig. 21. Influence of pH of dye solution on decolorization efficiency of Acid Blue dye.

terminal surface with a resulting lessening in the measure of shading oxidation.

# *4.5. Effect of pH of dye solution*

Solution pH is significant feature that effects on the competence of the plasma discharge process. To inspect its consequence, the pH of the samples was adapted to the determined value by addition sulfuric acid or sodium hydroxide solution using the 100 ml volume of solution, operation voltage15 kV, room temperature and pressure and initial concentration 10 ppm. The influence of original pH on the decolorization efficiency was obtained in Fig. 21. This figure indicates that decolorization proportion diminished gradually with the expansion at pH at the studied range of 3-6. For pH values above 9, the dye decolorization efficiency reduces quickly. The greatest color decolorization of 97.71% was recorded at pH = 3, and the lowest dye decolorization of 38.16% was recorded at  $pH = 11$ . With increasing solution  $pH$ , the adsorption limit for each concentration was decreased. This behavior might be returned to the anionic nature of the acid



Fig. 22. Relation between the corona current and the conductivity with different pH of Acid Blue dye solution.

blue dye that might be neutralized at the acidic medium that upgrades the decolorization procedure. While, at the basic media the anionic atoms of colored dyes was kept up contrarily charged. Fig. 22 displays the relation between the crown current under different pH values. It was indicated that corona current was decreased from pH 3 to pH 7 then ascents with expanding the pH until pH 11. Corona current is related to conductivity and ionic versatility, which is more noteworthy for the littler particles. The smaller the pH, the more prominent the corona current. As solution pH increments above 6, the increment at the current is frequently attributable to the including of NaOH utilized as base. The conductivity of Na<sup>+</sup> is high. In this manner, the corona current comparatively rises when the pH rises, yet lower. Also, there is a corona current least at neutral solution pH.

# **5. Conclusion**

This article deals with model for double pin arrangement in 3D program that was utilized for field and voltage. The simulation of non-thermal corona discharge plasma that can be clue to a superior considerate of the mechanism complex and can be utilized in the optimization of devices. The air distance for more corona region that can produced more ozone and active radicles that help in water treatment was optimized. In this investigation, the treatment by the release of the voltage in solution of Acid Blue80 dye was approved out and the color removal efficiency was calculated as 80% color removal by ozonation. The total color removal was succeeded at time 35 min for an original concentrations range of 5 ppm up to 100 ppm.The influence of several working factors such as original concentration, contact time, solution pH and gap distance on the competence of dye removal was examined. The outcomes presented that, greatest gap separation for that arrangement of 15 kV is 5 mm, the decolorization rate rise with the rise in contact time, and it was initiated to be reduced with the improvement at the initial concentration. The pH was found to have important influences on the rate of oxidation by corona discharge of the dye in the pH range from 3 to 11. It was specified that the best pH for the color removal of

acid blue dye is in the range between 3 and 6. Moreover, the corona current was measurement at each value of solution pH. Furthermore, the discharge energy and corona current were measured at different gap distances. Energy yield and electrical energy per order (EE/O) for dye decolorization process under diverse gap distances were calculated.

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