ISSN: 2350-0328

International Journal of Advanced Research in Science, Engineering and Technology

Vol. 7, Issue 10 , October 2020

Generation of High Voltage Discharge Plasma for Wastewater Treatment

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ABSTRACT: This work presents the generation of discharge plasma using a high voltage ac source in theair at atmospheric pressure above the water surface. A point-plane electrode configuration used to have different air gap distance conditions where positive electrode being placed in the atmospheric air at 1 mm and 5 mm gap distance from the water surface while the ground electrode remain submerged in water solution at a gap distance of 30 mm gap in all cases. It is observed that the discharge generation and its properties are significantly influenced by varying gap distances above the water surface. The plasma induced chemical effects of discharges in the reactor is evaluated: (i) by the measurement of solution conductivity and pH and (ii) by the treatment of methyl orange dye solution having different air gap distances. The degradation results of methyl orange dye are found comparatively faster for 5 mm gap distance than those for 1 mm gap distance. The experimental results indicate that the electrode gap distance from positive electrode tip to the water surface is an important factor that affects the physical and chemical processes of the high voltage discharge in reactor. Thus, the generation of high voltage discharge plasma can be used as an effective treatment method for industrial in general and textile wastewater in particular.

KEY WORDS: Discharge plasma; high voltage; gap distance, water surface, air, streamer, spark-like, methyl orange.

I. INTRODUCTION

The problems of wastewater become a serious environmental issue facing the world, as all living beings including humans are heavily dependent on water for food production and other purposes [1-2]. Everyday various kinds of organic and inorganic pollutants are discharged in most water bodies from textile, fertilizer, drug or chemical factories, agricultural lands and also from city sewage. Dyeing section of textiles contributes to 15%-20% of the total wastewater flow [2-4]. Over the past few decades, various physicochemical and electrochemical methods such as UV photolysis, photocatalysis, sonochemistry, supercritical water oxidation, have been examined for the removal and degradation of dye molecules from wastewater [1-10]. However, these methods suffer inherent disadvantages in terms of their applicability and costs. Therefore, the development of an advanced dye wastewater treatment method is very important to control water pollution. Recently a high voltage generated discharge plasma in or above water is considered as an effective wastewater treatment method [5-7]. The application of a high voltage across the electrode is able to concentrate a strong electric field at the high voltage needle leading to an easy breakdown of water to initiate discharge. The discharge can be in the form of corona or streamer, spark and arc make conductive channels producing high energy electrons in water which are capable of ionization, dissociation and/or recombination of water molecules [5-7]. Through these processes, a discharge plasma interacts with water molecules causing various physical and chemical processes in water such as a strong electric field (EF), intense UV radiation, shockwaves, and the generation of various active ions such as H^+, H_3O^+, O^+, H-, O-, reactive radicals such as OH_2, O_2, OH; and molecular species such as H_2, O_3, H_2O_2 [5-10]. These chemically active species generated by the electrical discharge can attack and then degrade the organic pollutants contained in the water. The development of streamers and their propagation depend on many factors including propagation medium, solution conductivity, pulse duration, pulse repetition rate, heating, pressure, input power, electrodes geometry, gas-liquid interfacial contacting patterns, and other plasma properties [6, 11-16]. Several of these factors have been studied by researchers, although many of their effects on water surface discharge are not yet well understood.

Y. Hayashi et al reported the decoloration of methyl orange dye using pulsed high-voltage discharge plasma generated in argon atmosphere at 313 K [8]. Different experiments were conducted using Copper (Cu) and stainless steel #304 (SUS-304) in the batch reactor. They observed methyl orange degraded into its derivatives with a change to a light

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International Journal of Advanced Research in Science, Engineering and Technology
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colorafter application of pulsed discharge plasma. The intermediate compounds formed from the degradation of methyl orange consisted primarily of aromatic compounds containing nitrogen functional groups. At the same number of plasma discharge times, the decoloration of methyl orange was lower with SUS than that with Cu as the electrode.

M. Sayed presented the effect of solution pH, temperature, treatment time on the degradation of phenol by pulsed high-voltage discharge [9]. He found the phenol degradation efficiency was increased with the increase of initial solution pH, temperature and treatment time. When hydrogen peroxide was added to the solution at a concentration of 5.3 mM, 10.6 mM or 20 mM, the phenol removal efficiency increased dramatically.

M. Sato et al studied the gas phase discharge generated on water surface using high voltage pulsed generator with 0-30 kV at 100 Hz pulse repetition frequency [10]. They found that the discharge characteristics varied with varying electrode distance between needle and water surface, and also with variation of ground electrode shapes. The discharge state changed from streamer mode to spark mode with decreasing electrode distance; discharge shape varied to circle, semi-circle and straight with the corresponding ground electrode shapes.The light intensity of discharge found to be brighter with increasing the supply voltage. Phenol was treated as wastewater sample using argon, air and oxygen as gas medium above water surface. It was found that the decomposition rate of phenol better in case of the oxygen than the argon and air.

M. I. Nawaz et al described the dielectric barrier discharge (DBD) system for the degradation of nitrobenzene (NB) in water [11]. They compare the degradation results from DBD system with advanced oxidation process (AOP) and found the faster degradation of NB using DBD than AOP. More ozone concentration was measured with increasing oxygen gas flow rate in DBD reactor which influence the degradation rate of NB. Also, the degradation of NB was significantly improved by increasing the voltage, gas flow rate, while it was decreased with the addition of inhibitors and also decreased with the presence of methanol.

N. K. Permatasari et al showed treatment method of tofu wastewater using contact glow discharge electrolysis (CGDE) with variation of air injection flow rate [12]. The experimental results showed that the production of H_2O_2 was higher during the CGDE process. It was also showed that in addition to air injection, the resulting specific energy is lower. With increasing the addition of air injection chemical oxygen demand(COD) degradation was higher.

The main purpose of this research is to provide a fundamental understanding of the generation processes of a discharge plasma in air at atmospheric pressure to water surface by a high voltage test transformer; where the performance of this discharge process is evaluated by the treatment of wastewater sample i.e. degradation of methyl orange (MO) organic dye with varying positive electrode gap distances to water surface.

II. EXPERIMENTAL SET UP

A block diagram of the experimental setup is shown in Fig.1. A step-up transformer (Model No. HV 9000, TERCO, 50Hz, rated secondary voltage: 140kV) consists of three windings with insulating shell and corona free aluminium shielded electrodes is used as high voltage supply source. A control desk is used to control and operate supply voltage across the positive and negative electrodes of a discharge reactor. The control desk contains all the control elements for operation of high voltage test equipment and measuring instruments. The control desk includes a motor operated regulating unit, consisting of ring-core regulating transformer and an isolating transformer. A capacitor of 1000 pF is charged with the conducting rod and energy charged by the capacitor is released to the electrodes. High voltage and current changes throughout discharge propagation are recorded by voltmeter and ammeter in control panel.The point-to-plane electrode configuration consists of a tungsten wire of approximately 0.5 mm diameter with a sharp tip being used as a positive electrode, andan aluminium plate of 20 mm diameter and 0.2 mm thicknessas a ground placed in a cylindrical glass reactor. For different experiments, the positive electrode is placed in atmospheric air at 1 mm and 5 mm gap distance above water surface, while in both cases theground plate is immersed in water at 35 mm from the watersurface. The sample solution is prepared at 100µS/cm dissolving NaCl in pure distilled water. The conductivity of water solution is measured using a conductivity tester (Lutron, PCD-431) and pH is measured using a pH meter (Lutron, PH-208). Methyl orange dye ($C_{14}H_{14}N_3NaO_3S$) is used to prepare the wastewater sample which is widely used in textile industries. The absorption spectra of dye are measured using an absorption spectrophotometer (model: 721D) at 560-

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565nm. For preparation of the dye solution, 0.5 g dye powder is mixed with 1 L of distilled water and then the conductivity is adjusted using required NaCl. The volume of treated solution used for the experiments is 200 mL.

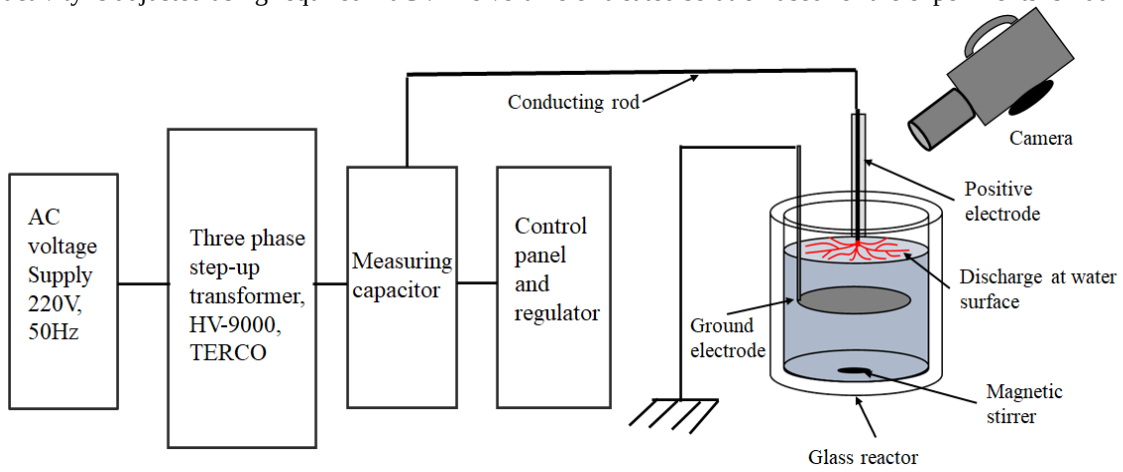

Fig. 1: Experimental setup for generation of high voltage discharge in the atmospheric air to water surface.

III. RESULTS AND DISCUSSIONS

(a) CASE 1: GENERATION OF HIGH VOLTAGE DISCHARGE AT A GAP DISTANCE OF 1MM

In this case, the positive electrode tip is placed at 1 mm gap in the atmospheric air from the water surface and ground electrode is immersed in water solution at 35 mm distance from water surface. When the high-voltage is applied across the electrodes, the discharge is initiated from the needle tip propagating through air gap to the water surface. It is observed that the discharge initiated at 1.64 kV is of a very low light intensity having only 0.1 A conduction current at 0.24 second. But with increasing the supply voltage, the discharge size increases with bright light intensity. The variation of voltage and conduction current during discharge development are shown in Table I. It reveals that the conduction current reaches 12.90A during the full development of the discharge. At this time several streamer branches are observed over the water surface. Typical images of discharge development with time are shown in Fig. 2. It is seen that the discharge size and light intensity are very low at 0.48 second and then spread over the water surface at 0.50 second having thin streamer branches. Finally, it reaches the full development at 0.51 second, and at this time conduction current reaches 12.90 A. For the full development of the discharge, the required supply voltage is 6.98 kV. Due to full development the voltage suddenly decreased to 2.97 kV at 0.51 second. In this case discharge is characterized as only streamer with thin branches and their length cannot touch the ground electrode of the reactor.

Table I: A typical change of voltage and conductive current during propagation of discharge at 1 mm gap distance

Time (sec)	Current (A)	Voltage (kV)
00:21	0	0.74
00:22	0	0.80
00:24	0.1	1.64
00:25	0.1	1.82
00:44	0.2	5.50
00:46	0.3	5.75
00:48	0.3	6.08
00:49	0.3	6.45
00.50	12.50	6.98
00.51	12.90	2.97

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00.51	0.50	1.64

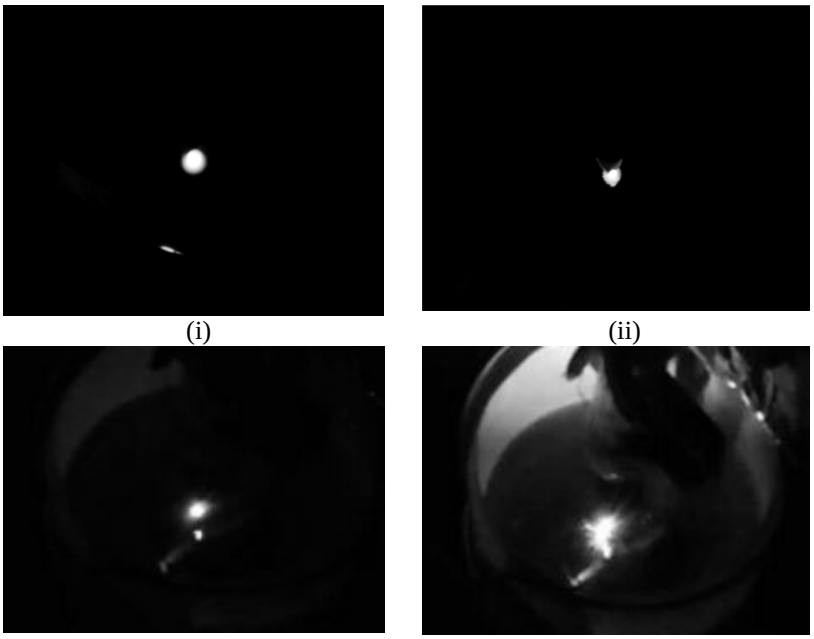

Fig. 2: Typical images of high voltage discharge at water surface when electrode gap distance is 1 mm from positive electrode tip to water surface: (i) at 0.48, (ii) at 0.49, (iii) at 0.50 and (iii) at 0.51 second

(b) CASE 2: GENERATION OF HIGH VOLTAGE DISCHARGE AT A GAP DISTANCE OF 5 MM

In this case, the positive electrode tip is put at 5 mm gap in the atmospheric air above the water surface whereas ground electrode is immersed at the same distance from the water surface. It is observed that the discharge initiated at 1.82 kV of supply voltage at 0.31 second with conduction current is 0.1 A. After that it crosses the air gap immediately with a long streamer length and then spreads over the water surface with several thin streamer branches. The variation of voltage and conduction current during discharge development is shown in Table II. It is seen that the conduction current increases very sharply from 13.50 to 18.40 at 0.40 to 0.44 second, respectively. However, the required supply voltage is 3.93 kV for the full development of the discharge, which is lower than that at 1 mm gap distance case. Typical images of discharge development with time is shown in Fig. 3. It is observed that the discharge crossing the air gap with a low light intensity at 0.37 second, then discharge become stronger with bright light intensity at about 0.40 second and spreads over the water surface with several streamer branches at 0.41 second. After the full development of the discharge branches are long enough to touch the ground electrode and finally discharge converted from streamer branches to spark-like discharge at 0.44 second.

From comparison of case 1 and case 2, it is clear that the physical characteristics and propagation process of discharge are significantly varied with a varying the gap distance from 1 mm to 5 mm above the water surface. Moreover, conductive current is much higher at 5 mm than that at 1 mm gap distance. It means that electric field is much stronger at 5 mm gap distance condition which may lead to increasing ionization of air and water upon plasma contact during propagation of discharges.However, transition of discharge characteristics depends on the electric field condition that is formed by high voltage supply and ionization of surrounding medium of the electrodes [2, 14-17].

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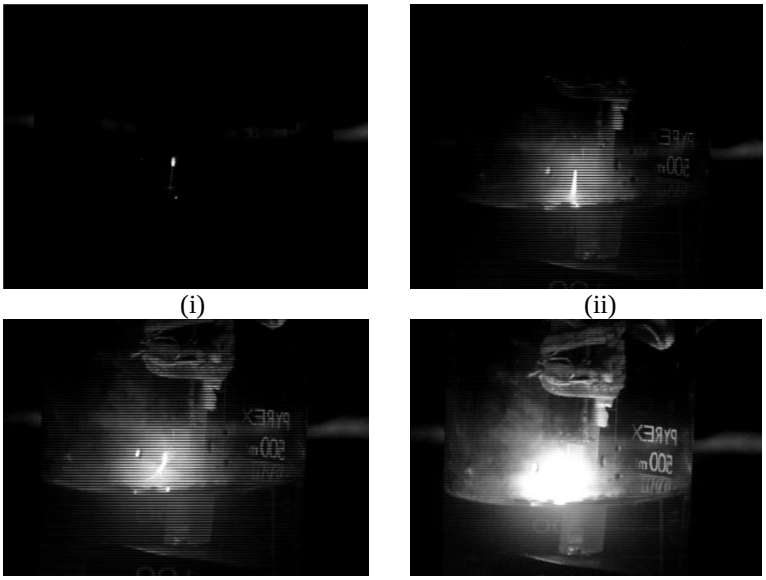

Fig. 3: Typical images of high voltage discharge at water surface when electrode gap distance is 5mm from positive electrode tip to water surface: (i) at 0.37, (ii) at 0.40, (iii) at 0.41 and (iii) at 0.44 second.

Table II: A typical change of voltage and conductive current during propagation of discharge at a gap distance of 5 mm

Time (sec)	Current (A)	Voltage (kV)
00:30	0.0	1.55
00:31	0.1	1.82
00:32	0.1	2.20
00:37	0.2	3.74
00:40	13.50	3.93
00:41	12.80	2.11
00:42	10.10	2.00
00.43	11.50	2.57
00.44	18.40	1.80
00.47	9.50	2.84

(c) MEASUREMENT OF SOLUTION CONDUCTIVITY AND pH AT DIFFERENT GAP DISTANCE CONDITION

The measurement of solution conductivity and pH is performed with a varying of gap distance to understand the physical effects of discharge on plasma chemical activity in discharge solution in the reactor. Typical variation of (a) conductivity and (b) pH of water solution by propagation of high voltage discharge from air to water surface at 1 mm and 5 mm gap distance condition is shown in Fig.4. It is found that conductivity increases from 100 to 169 µS/cm and 109 to 290 µS/cm at 1 mm and 5 mm gap distance condition, respectively. On the other hand, the pH value decreases from 5.98 to 4.67 and 6.52 to 3.8 at 1 mm and 5 mm gap distance, respectively. From above discussions and results it is clear that chemical properties of discharge mostly influence by the physical processes of discharge in the reactor. It is expected that strong physical processes such as a high electric field, UV light emission, heat generation happening at 5

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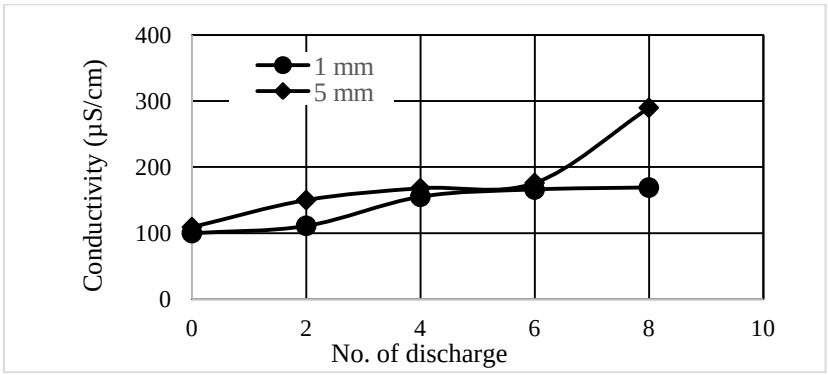
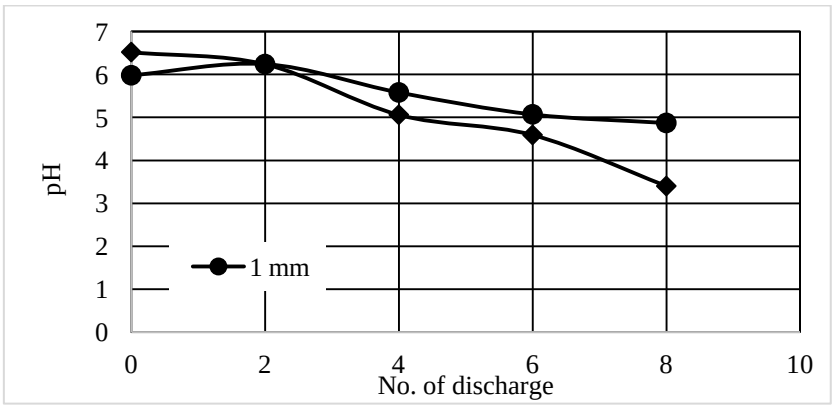
mm gap than that at 1 mm gap distance condition. As a result, far more formation of various ions and radicals might be taking place during the propagation of discharges at 5 mm gap than that in case of 1 mm gap distance.

In addition, the large area of water surface possibly comes into contact with discharge channels due to its full development at 5 mm gap distance which helps to produce more chemical species in the discharge reactor compared to 1 mm gap distance. Moreover, discharge channel head probably contains more high energetic electrons at 5 mm gap which causes longer discharge lengths at water surface. For this, discharge type might be easily converted to spark like discharge resulting in increased production of chemical species in the discharge reactor.

High voltage discharge on water surface produces a variety of reactive radicals (OH, O, H, HO_2^-), ions (H^+, O^-, H_3O^+) and molecular species (H_2, O_2, O_3, H_2O_2) during their propagation from air to water surface [7-9, 14-16]. After initiation, when the streamer directly propagates through air or gas phase from the high voltage needle tip, the primary radicals or ions are expected to be generated in air or air-water interface by the contact plasma. Then these are injected to water which subsequently react with water molecules on surface to form secondary molecular species [16]. It takes some time for the active species to diffuse from the air plasma to the water surface. During that time, many of the primary radicals probably disappear before reaching the water surface due to their short lifespan. The active species which diffuse in the air phase and dissolve in the water through the surface layer to react with the organic materials or microorganisms in water [13-14].It is suggested, among these active species, hydroxyl radicals (OH·) and hydrogen peroxide (H_2O_2) are the most important chemical species taking part in the breakdown of the organic contaminant degradation due to their high oxidizing power [13-14]. In the vapor phase or in air-water interface OH· radicals could be generated by electronic or thermal dissociation of water molecules upon contact with the plasma prior to recombining into H_2O_2 which quickly dissolves in water [13-16].



(a)



(b)

Fig. 4: Typical variation of (a) conductivity and (b) pH of water solution by propagation of high voltage discharge from air to water surface at 1 mm and 5 mm gap distance condition.

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(d) TREATMENT OF WASTEWATER SAMPLE (METHYL ORANGE DYE SOLUTION) AT DIFFERENT GAP DISTANCE CONDITION

This research aims to apply a high voltage discharge for wastewater treatment to understand the plasma induced chemical processes produced by physical properties of discharge (streamer or sparklike) at 1 mm and 5 mm gap distance condition. Here methyl orange ($C_{14}H_{14}N_3NaO_3S$), an organic dye is selected as a model wastewater sample, which is one kind of azo dye and widely used in textile sectors [17-19]. This dye is characterized by an azo group consisting of two nitrogen atoms (-N = N-), which is very sensitive to OH· radicals. Dye degradation can be easily monitored by measuring the sample of the dye color at the wavelength of absorption (λ = 560-565 nm) using a spectrophotometer [17-19]. For this, degradation rate of methyl orange dye is evaluated with a varying gap distance between positive electrode tip in air to water surface in the reactor.

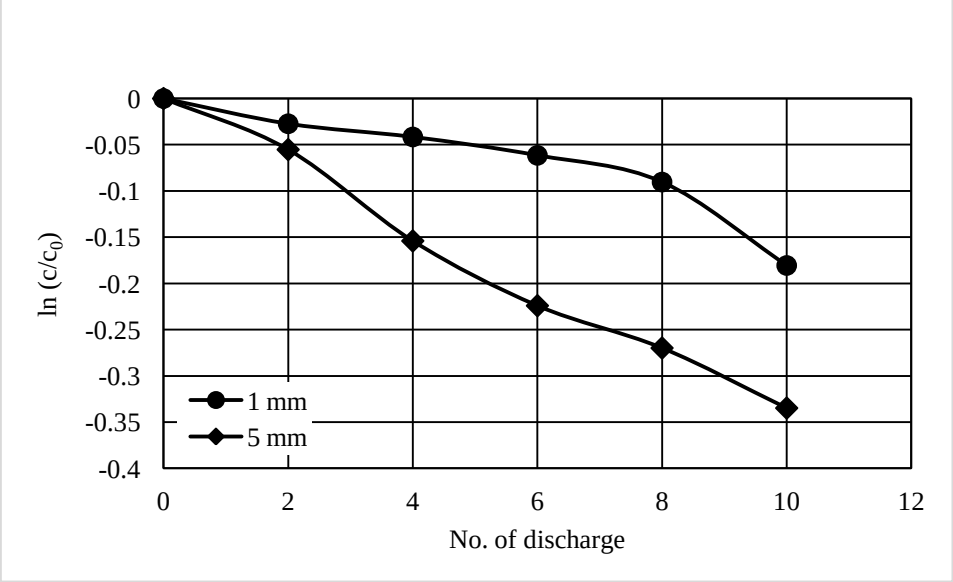

Fig. 5: Comparison plot of degradation of methyl orange dye using high voltage discharge from the air to the water surface at 1 mm and 5 mm gap distance condition.

Experimental results on degradation of methyl orange at 1mm and 5 mm gap distance are shown in Fig. 5. The degradation rate estimated from the curve fit was 1.59E-02 and 3.00E-02 at 1 mm and at 5 mm gap distance, respectively; which is about 53% faster at 5 mm than that at 1 mm gap distance. These results indicate a strong dependence of plasma chemical activity happened by physical properties of discharge plasma from the atmospheric air to water surface. From above discussion in section (a) and (b), we see that discharge is initiated with a very low light intensity; propagated with small length thin branches over the water surface at 1 mm gap distance. During this period of time the conduction current consumed by discharge is found lower. This may be due to a small amount of air get ionized in the air because of 1 mm gap distance above the water surface; resulting in fewer chemical species on water surface than that at 5 mm gap. As a result, fewer reactive species may be produced in the air and air-water interface. Also, a small area of water surface comes in contact with discharge plasma causing the formation of decreased chemical species such as OH· radicals. Therefore, only small amount of methyl orange dye molecules is able to break down. On the other hand, as the discharge crosses the air gap with a longer length in the air due to 5 mm gap distance above the water surface, a large amount of air gets ionized; leading to increased reactive species being produced in the air and air-water interface; forms more chemical species at the water surface than these at 1 mm gap. Under this condition the conduction current consumed by the discharge is higher. Discharge propagated strongly with longer length branches over the water surface. Because of this, a wide area water surface comes in contact with the discharge plasma, Thereby, more reactive water molecules also come in contact with the discharge branch leading to a higher amount of chemical species formed at the plasma zone during the propagation of discharge. In addition, conversion of discharge from streamer branches to sparklike at 5 mm gap probably helps to produce more chemical species in the

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reactor resulting in more breakdown of methyl orange dye molecules. For this reason, degradation of methyl orange dye is faster at 1 mm than that at 1 mm gap distance. Moreover, it is seen that UV radiation generated from spark like discharge is much stronger for 5 mm than 1 mm gap condition. To confirm chemical activity of OH· radicals on degradation of methyl orange, an experiment has been conducted experiment with an addition of H_2O_2 to dye solution with and without heating in the laboratory in absence of any discharge. This is because H_2O_2 could be formed in the discharge region by the combination of OH· radicals and also the dissociation of H_2O_2 gives OH· radicals in water [14-16]. The experimental result including this effect is shown in Figure 6, where it is found that degradation rate is low with the addition of H_2O_2 only (10 drops) at ambient temperature; however, degradation rate improves significantly in the experiment keeping solution temperature at 30°C and 40°C with the same amount of H_2O_2. The temperature of the reactor is kept constant using a stirring and heating machine. It is proved that an increase in temperature causes increased production of OH· radicals by dissociation of H_2O_2 in dye solution. As a result, degradation rate is found higher at 40°C than that at 30°C with the same amount of H_2O_2. Degradation rates from curve fit of Fig. 6 are 1.6E-03, 3.2E-03 and 3.8E-03 under the conditions: (i) at ambient temperature with only H_2O_2, (ii) at 30°C with H_2O_2 and (iii) at 40°C with H_2O_2 respectively.

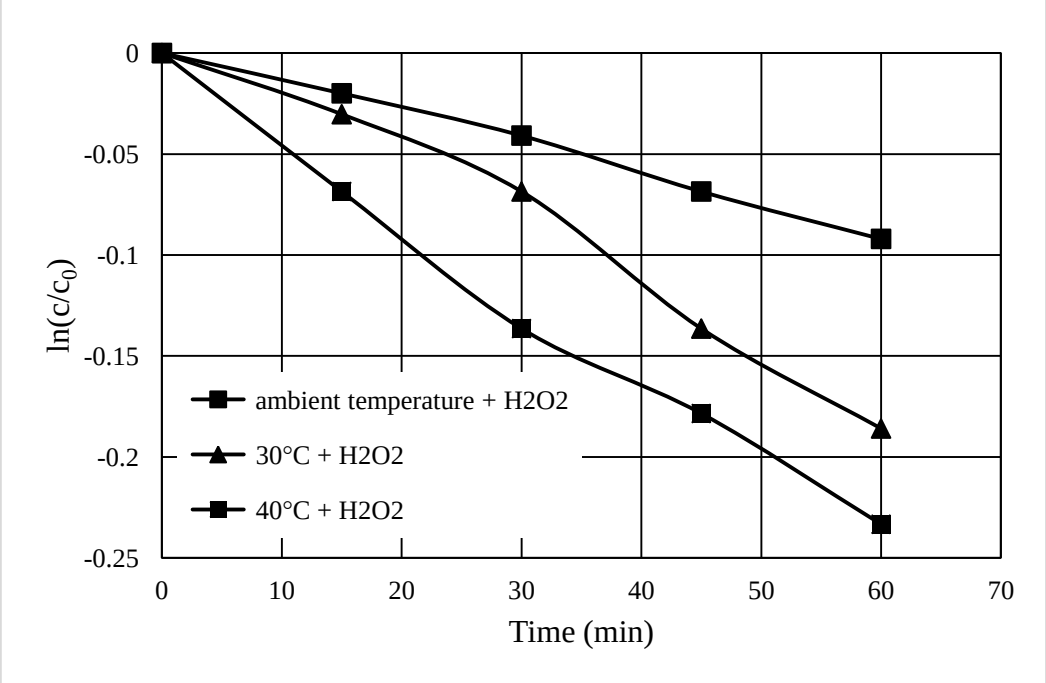

Fig. 6: Comparison of degradation of methyl orange dye treated under the condition: (i) at ambient temperature with H_2O_2, (ii) at 30 °C with H_2O_2 and (iii) at 40 °C with H_2O_2.

IV.CONCLUSION

The discharge plasma is generated successfully at 1 mm and 5 mm gap distance in the atmospheric air to the water surface by high voltage ac supply. A three-phase step-up transformer (HV-9000, TERCO) is used as a supply source. For all experiments, the initial conductivity of water solution is 100 µS/cm. It is seen that a comparatively weak and small size discharge generated at 1 mm gap distance and finally discharge is characterized as streamer with thin branches. However, under 5 mm gap distance condition, a higher voltage is required for the initiation of discharge, that crosses the air gap with a long streamer channel with bright light intensity; and then the streamer branches spread over the water surface with longer length and finally it converted from streamer to sparklike discharge. The solution conductivity and pH are measured from discharge water as indicator of chemical species being formed in the reactor

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through discharge processes. Methyl orange organic dye solution is used in both cases to clarify the plasma induced chemical activity of the discharge in water solution.

The experimental results of methyl orange (MO) dye treatment have shown that degradation rate of dye solution is faster at 5 mm than 1 mm gap distance case, which is in good agreement with physical and chemical properties resulted from the discharge at water surface. To confirm the role and effect of OH· radicals and temperature generated by discharge on the degradation of methyl orange, experiments are conducted using only H_2O_2 without any discharge. It is found that the degradation rate is very negligible with H_2O_2 only, degradation rate is found to increase as the solution temperature is kept constant at 40° C than 30° C with same amount of H_2O_2. From the above results and discussion, it is clear that both physical and chemical effects generated by high voltage discharge in the reactor play an important role in the removal of organic pollutants from wastewater. So, the generation of high voltage discharge plasma turns out to be an effective method for the treatment of industrial and especially textile waste water.

ACKNOWLEDGEMENTS

This work was supported by University Teacher Research Fund of University Grants Commission of Bangladesh (UGC). The author would like to thanks to all the committee members of CASR, Dhaka University of Engineering & Technology (DUET), Gazipur for selecting this research to support. Special thanks go to the Head, Department of Electrical and Electronic Engineering (EEE) and the Director of Research & Extension for their continuous support during this research.

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