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# Development of Multi-Compartment Dielectric Barrier Discharge Plasma Reactor for Innovative Water Treatment

Umesh Chavan[1*], Dattatray Hulwan[1], Anuj Harad[1], Arya Gham[1], Prathamesh Bagale[1], Swapnil Aphale[1], Swaraj Ajgaonkar[1]

[1] Department of Mechanical Engineering, Vishwakarma Institute of Technology, 411 037 Pune, Maharashtra, India
[*] Corresponding author, e-mail: umesh.chavan@vit.edu

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**Abstract**

A novel multi-compartment dielectric barrier discharge (MCDBD) plasma rector is developed and tested to produce plasma-activated water (PAW). MCDBD reactor consists of a polycarbonate container with six compartments. The top electrodes are stainless steel needles connected to AC high-voltage power supply. The bottom of each compartment is replaced with a glass slab and stainless-steel mesh electrodes. Cold plasma is generated in all compartments simultaneously to activate water. Experiments conducted by varying treatment time, power levels, gap between electrodes and water surface, and volume of water. Production of reactive oxygen and nitrogen species (ROS and RNS, respectively) in PAW is evaluated as per international standards (APHA/AWWA/IS). The application PAW greatly depends on the ROS and RNS concentration so results of MCDBD reactor are compared with conventional reactors. ROS and RNS concentration in 1800 mL water is measured 0.93 M and 0.52 M, respectively, in the MCDBD reactor. ROS and RNS concentration in 500 mL water is found to be 0.6 M and 0.44 M, respectively, in a conventional reactor. Result shows higher concentration of ROS and RNS produced in large volumes of water using MCDBD reactors. Even though the same amount of power is supplied to both conventional and multicompartment reactors, output in terms of ROS and RNS production is significantly greater in new design. Also, ROS and RNS have longer life in MCDBD reactor which is desirable to deactivate the biofilms and water decontamination. Proposed design is found to be more suitable for wastewater treatments, biomedical and agriculture applications.

**Keywords**

dielectric barrier discharge, plasma reactor, reactive oxygen and nitrogen species, plasma activated water

## 1 Introduction

As a reaction to the ever-increasing human population on the planet, there is an increasing demand for drinkable water which has pushed researchers to research on various treatments possible for water from industrial wastes, fresh but contaminated sources and even seawater [1, 2]. The current treatments are primarily of three types: physical, biological and chemical out of which chemical is the one which is majorly used in industries for secondary treatment of industry waste water. Physical treatments are generally employed in primary treatment of water whereas chemical and biological treatments are employed in secondary treatments [3, 4]. Some of the physical treatments might include adsorption, coagulation, floatation, foam fractionation, reverse osmosis, etc. Chemical treatments include electrolysis, ion exchange, oxidation, neutralization, reduction, etc. Whereas flocculation, fungal treatment, stabilization, aerated lagoons, activated sludge, trickling filters come under biological treatments [5, 6]. However, these processes possess some disadvantages which limit the extensive use of these processes such as no single unique method to reduce compounds like heavy metals, nitrogen, phosphorus, etc. New techniques like advanced oxidation processes (AOPs) are being researched in recent times to overcome these types of issues. Researchers have studied advanced oxidation processes wherein they included UV/$O_3$, UV/$H_2O_2$, photo-Fenton, Fenton, non-thermal plasmas, radiolysis, supercritical water oxidation processes, etc. This set of oxidative water treatments that are used to treat wastewater is what constitutes an advanced oxidation process. Some of the advantages of using these AOPs are their high reaction rates, not creating a sludge is seen

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when physical, chemical or biological processes, potential to reduce toxicity, mineralize the organic contaminants and low cost [1, 7, 8]. Strong electric fields when immersed in water (called electro hydraulic discharge) creating plasmas can have impacts that both physical and chemical treatments have on infected water; and it is this application of plasmas that researchers are attracted to in recent years [9, 10]. The high-voltage power supply characteristics of high-voltage discharge for plasma generation should be such that the voltage pulse should quickly reach its peak and the duration of the pulse should be short [11, 12]. When non-thermal plasma producing probes rotating at 1000 rpm were submerged in a solution containing then a 6-log reduction in their colonies is noticed after a time period of 20 and 40 s, respectively [13, 14]. Another study [15–19] suggested that the SARS-CoV-2 virus can be deactivated by submerging arc discharge plasma reactor by forming various reactive nitrogen species and other types of chemically active species. Also, as reported [20–22] the pulsed corona discharges when burned in oxygen and the liquid solution is sprayed in the plasma affecting area gives the most efficient reactors in terms of energy yield. Now is also the right time to discuss the types of plasma generation used specifically for water treatment. In total there are many methods for plasma generation. Some of them include: dielectric barrier discharges (DBDs), contact glow discharges, spark discharges, gliding arc, corona discharges etc. Among these DBD and corona discharge types are the most researched ones [23–25]. Previous studies indicate that dielectric barrier discharge (DBD) methods are effective for water treatment because they provide a more homogeneous discharge compared to volume dielectric barrier discharge (VDBD) geometries [26–29]. However, the treatment of large volumes of water and treatment efficiency is a challenge. The long-lived chemically active species, especially hydrogen peroxide, ozone and nitrate ions which are produced by a water film DBD reactor, were measured. The contribution of long-lived active species to p-nitrophenol degradation in the falling film membrane reactor was evaluated [30–32]. The synergetic degradation performance of Acid Orange 7 (AO7) by dielectric barrier discharge plasma (DBDP) and persulfate (PS) was examined [33]. This study investigates the application of a dielectric barrier discharge plasma reactor for water treatment. A novel multi compartment dielectric barrier discharge (MCDBD) plasma rector is developed and tested to produce plasma activated water. The experiments were conducted varying treatment time, power levels, gap between electrodes and water surface and volume of water. Production of reactive oxygen and nitrogen species (ROS and RNS, respectively) in plasma activated water are evaluated as per international APHA/AWWA/IS standards [34–36]. Results of MCDBD plasma reactors are compared with conventional plasma reactors. Thus, this research might provide an important piece of experimental study done on the subject matter.

## 2 Design of multi-compartment DBD plasma reactor

DBD type reactors are more popular to produce ROS and RNS in water. DBD configuration includes direct and indirect discharge of plasma, also, discharging the plasma in bubbles on the surface of the conducting fluid or water. When plasma is used for water treatment, the high number of electric collisions produced due to high electric field has a physical impact on the contaminated water, whereas on molecular level an array of ionic, molecular compounds as well as different types of chemically active species are produced. This is how plasma discharge is used for virus and bacteria deactivation, chemical degradation and removal of contaminants from water. In present research plasma generator design is based on DBD principle as shown in Fig. 1. MCDBD plasma reactor consists of a polycarbonate plastic container with six compartments shown in Fig 1. Plasma can be discharged in all compartments simultaneously to treat the water. The top electrodes are stainless steel needles connected with alternating current (AC) high voltage power supply. The bottom of each compartment is replaced with a glass slab and stainless-steel mesh electrodes and this forms ground electrodes at the bottom of the compartment as shown in Fig. 1. Each compartment has a capacity of 500 mL; however, water treatment testing is carried out for different water to air volume ratios. The MCDBD plasma reactor proposed in this research ensures separating the water using six compartments and setting electrodes

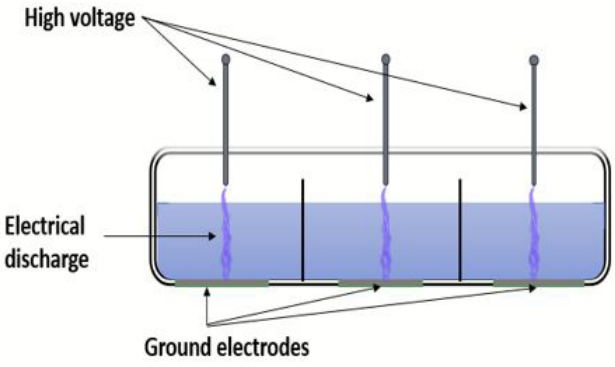


**Fig. 1** Working principle of MCDBD plasma reactor

for each compartment. Here, it is possible to maintain the high plasma discharge density in each compartment and treat a large volume of water. MCDBD reactors actively produce highly reactive species which destroy the pollutants and biofilms in the vicinity of it efficiently.

Plasma produced by MCDBD reactor is non-thermal in nature, there was no cooling system needed in actual implementation. Surface dielectric barrier discharge (SDBD) concept is used to build reactors. In the proposed design, the plasma is produced at the surface of the dielectric, which assures a more homogenous type of discharge.

## 3 Water and plasma chemistry

The formation of ROS and RNS are described through chemical reactions (Eqs. (1)–(22), where $\sigma$ represents the wavelength range (in nanometers); M represents a third molecule that is present to absorb excess energy and stabilizes the reaction products (it is often an inert gas molecule, like $N_2$ or $O_2$, that does not chemically react but participates in the energy transfer process); and peroxynitrous acid (ONOOH) is a reactive nitrogen species [30–33]. These reactions are helpful to understand the main factors influencing the production of long-lived ROS and RNS and the role of these species in water decontamination. $O_3$ and $H_2O_2$ are the main active molecular ROS for water decontamination. There are two ways to form $O_3$ in gas–liquid phase electric discharge (Eqs. (1)–(3)) [32]. $H_2O_2$ is formed by the recombination of ˙OH radicals and reactions involving the production of hydrogen peroxide in gas–liquid discharge (Eqs. (4)–(8)) [30–33]. Electric discharge in air water interface results in formation of nitrite/nitrate species (RNS) elaborated through chemical reactions (Eqs. (9)–(22)) [30–33].

$$O_2(gas) + e \rightarrow O + O \quad (1)$$

$$O + O_2(gas) \rightarrow O_3(gas) \quad (2)$$

$$O_3(gas) \rightarrow O_3(aq) \quad (3)$$

$$e + H_2O \rightarrow e + H + {}^{\bullet}OH \quad (4)$$

$$H_2O + h\nu \rightarrow {}^{\bullet}OH + H \ (\sigma = 145 - 246 \text{ nm}) \quad (5)$$

$$OH + {}^{\bullet}OH \rightarrow H_2O_2 \quad (6)$$

$${}^{\bullet}H + {}^{\bullet}OH \rightarrow H_2O \quad (7)$$

$${}^{\bullet}H + {}^{\bullet}HO_2 \rightarrow H_2O_2 \quad (8)$$

$$N_2 + 2e^{-} \rightarrow 2N \quad (9)$$

$${}^{\bullet}N + 2{}^{\bullet}O \rightarrow NO_2 \quad (10)$$

$${}^{\bullet}N + O_3 \rightarrow {}^{\bullet}NO + O_3 \quad (11)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (12)$$

$${}^{\bullet}NO + O_3 \rightarrow {}^{\bullet}NO_2 + O_2 \quad (13)$$

$${}^{\bullet}NO_2 + {}^{\bullet}OH + M \rightarrow ONOOH + M \quad (14)$$

$${}^{\bullet}NO + {}^{\bullet}HO_2 + M \rightarrow ONOOH + M \quad (15)$$

$$ONOOH(aq) \rightarrow {}^{\bullet}NO_2(aq) + {}^{\bullet}OH(aq) \quad (16)$$

$$ONOOH(aq) \rightarrow HNO_3(aq) \quad (17)$$

$$H^{+}(aq) + NO_2^{-}(aq) + H_2O_2(aq) \rightarrow ONOOH(aq) + H_2O \quad (18)$$

$$NO_2(gas) \leftrightarrow NO_2(aq) \quad (19)$$

$$NO_2(aq) + H_2O \rightarrow HNO_3(aq) + HNO_2(aq) \quad (20)$$

$$HNO_3(aq) \rightarrow H^{+} + NO_3^{-} \quad (21)$$

$$HNO_2 \leftrightarrow H^{+} + NO_2^{-} \quad (22)$$

## 4 Experiments

The experimental layout shown in Fig. 2 elaborates the electronic devices used in experiments. AC power supply is connected to the witch-mode power supply (SMPS) which transfers power to the step-up transformer to convert the voltage to 12 kV–60 kV. This power is controlled by the Arduino uno which is then connected to the reactor

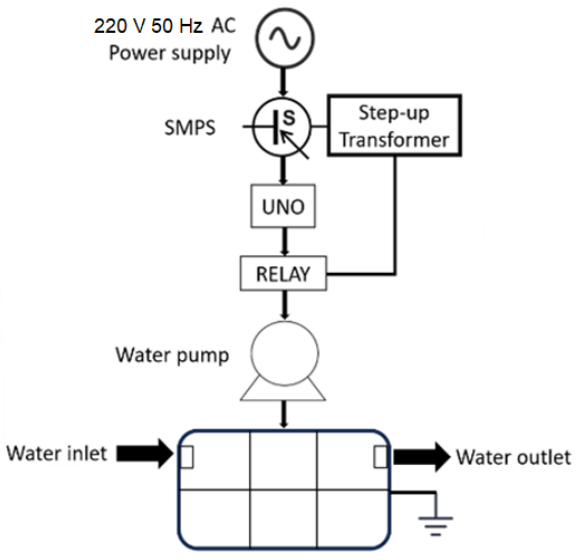


**Fig. 2** Layout of experimental setup (Plasma reactor)

via the relay module. Water pump is used for circulation of water across compartments to ensure uniform concentrations of ROS and RNS. Specifications of reactor and electric power details provided in Table 1.

The design of experiments (DOEs) shown in Table 2 elaborates the three variables considered in experimental study to investigate treatment efficacy. Namely volume of water, gap between electrodes and water surface, and treatment time. Experiments are designed to evaluate the effects of each variable individually and in combination on the treatment efficiency of the plasma system. Quantity of water used for treatment is 1 L, 1.5 L, and 1.8 L. The distance between the electrodes and water surface was varied, including 0.6 cm, 0.8 cm, and 1 cm. Finally, the treatment time was varied across three different levels, including 10 min, 15 min, and 20 min. As per design of the experiment table 45 experiments are conducted at a power level of 60 kV. Thus, at each level of power 45 experiments are performed.

The test results provided valuable insights into the optimal operating conditions for the cold plasma generator. The volume of water and gap between electrodes and water surface were found to have a significant impact on the plasma treatment efficiency. The optimal conditions for the volume of water and gap between electrodes are found to be 1.8 L and 1 cm, respectively. Fig. 3 and Fig. 4 shows actual water treatment process using MCDBD plasma reactor with six plasma jets. Water treatment processes is carried out as per DOE plan presented in Table 2. Fig. 5 shows the water treatment by single DBD plasma jet, and this type of plasma generator is referred as conventional plasma generator. 150 mL to 500 mL water is treated by single DBD plasma jet reactor. In all experiments ROS and RNS

**Table 1** Components used and their power supply characteristics

| Components | Specifications |
|---|---|
| Reactor | 3.2 L capacity, polycarbonate plastic |
| Electrodes and dielectric | Stainless steel (SS), 140 mm × ø 2 mm, 6 upper electrodes, ground electrodes of SS mesh, dielectric glass slab 1 mm thick |
| Pipelines | ø 10 mm silicon tubes |
| Power supply | 220 V, 50 Hz AC |
| Switch-mode power supply (SMPS) | 220 V AC supply, 5 A, 120 W, DC 24 V output |
| Pump | 12 V DC supply, 0.7 A, 7 W, 3 m |
| Transformer | Single phase, 12–60 kV, 1–5 kHz, step up |

**Fig. 3** Water treatment process using MCDBD plasma reactor (top view). Six compartments with DBD plasma jet in each compartment

**Table 2** Design of experiments (12 kV–60 kV)

| Experiment group number | Amount of water (mL) | Gap between electrode and water surface (cm) | Treatment time (min) | Number of runs | Number of total experiments |
|---|---|---|---|---|---|
| 1 | 1000 | 0.6 | 10 | 5 | 5 |
| 2 | 1000 | 0.8 | 20 | 5 | 5 |
| 3 | 1000 | 1 | 15 | 5 | 5 |
| 4 | 1500 | 0.6 | 20 | 5 | 5 |
| 5 | 1500 | 0.8 | 15 | 5 | 5 |
| 6 | 1500 | 1 | 10 | 5 | 5 |
| 7 | 1800 | 0.6 | 15 | 5 | 5 |
| 8 | 1800 | 0.8 | 10 | 5 | 5 |
| 9 | 1800 | 1 | 20 | 5 | 5 |
| Total experiments | | | | | 45 |

**Fig. 4** Water treatment process using MCDBD plasma reactor (front view). Height of reaction chamber and plasma jet

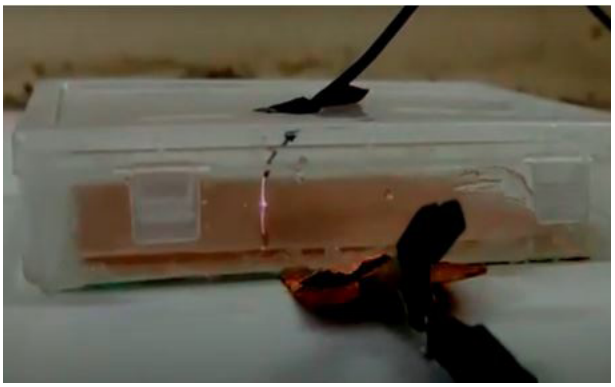

**Fig. 5** Single jet plasma reactor (conventional design)

concentrations in plasma activated water (PAW) are measured using standard test kit supplied by AQUASOL as per international standards (APHA/AWWA/IS) [34–36]. Plasma activated water testing is one the major activities of the current research. Reactive oxygen species measured in PAW are $H_2O_2$, $O_3$, and $O_2$. Similarly reactive nitrogen species measured in PAW are $NO_2$ and $NO_3$. Application of process and PAW greatly depend on concentrations of ROS and RNS. This technology has great potential and is suitable in wastewater treatments, bio medical, food industry, and agriculture applications [29–33].

## 5 Measurement of ROS and RNS

Concentrations of reactive oxygen and nitrogen species in plasma activated water are evaluated as per international standards [34–36] and classical chemical methods.

### 5.1 pH measurement with AQUASOL AE306 test kit

The standard test method is given in part 40 of IS 3025 [36]. The test result is shown in Fig. 6.

### 5.2 $O_3$ measurement with AQUASOL AEOZ3H test kit

Standard methods for chemical analysis of water and wastewater is described in method 4500-C1 F [34–36]. The


**Fig. 6** pH measuring with AQUASOL AE306 test kit, pH value (7.5 to 8)

comparison of the water samples of various ozone concentrations is shown in Fig. 7.

### 5.3 $H_2O_2$ measurement with AQUASOL AEHP test kit

Test standard follows the classical chemical methods. The comparison of samples without and with $H_2O_2$ content is shown in Fig. 8.

### 5.4 Dissolved oxygen measurement with AQUASOL AEDO8 test kit

Test standard follows the classical chemical method. The comparison of various samples is shown in Fig. 9.

### 5.5 $NO_2$ measurement with AQUASOL AE257 test kit

Standard methods for chemical analysis of water and wastewater is described in method 4500-NO2-B [34–36] and in part 34 of IS 3025 [36]. The result obtained with the test kit is shown in Fig. 10.

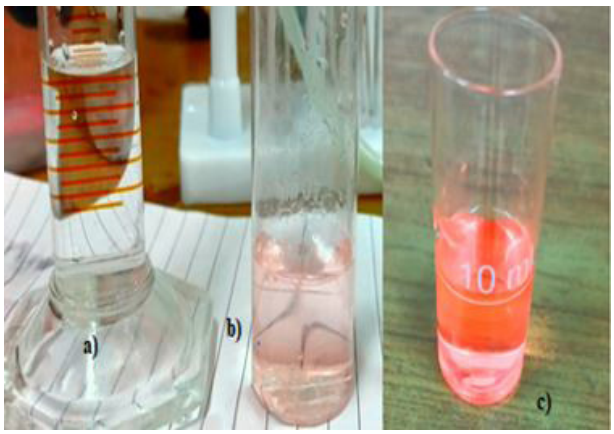


**Fig. 7** Ozone measurement a) Ozone test for untreated water (0 ppm) b) Ozone test for PAW (concentration 0.25 to 0.5 ppm), c) Ozone test for PAW (concentration 0.75 to 1 ppm)

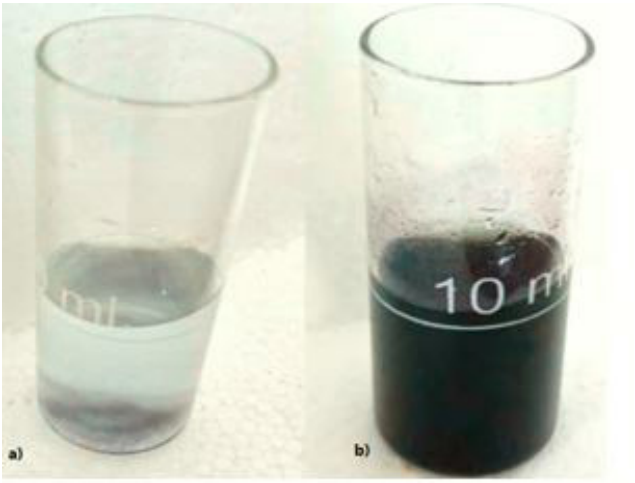


**Fig. 8** $H_2O_2$ measurement a) no change in color of sample (concentration 0 ppm) b) Black-brown color appeared $H_2O_2$ is present in sample (concentration 50 ppm)

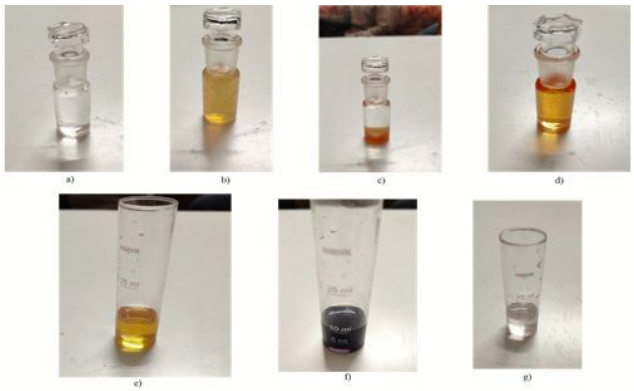

**Fig. 9** Dissolved oxygen (DO) measurement a) PAW sample for testing, b) After adding 10 drops of HP1 and 10 drops of HP2 solution to the sample, c) Sample after 20 min, d) Sample after adding of 10–12 drops of HP3 solution, e) 10 mL sample of water for further testing, f) Sample after adding 4 drops of HP4 solution, g) Sample after adding 13 drops of HP5 solution (DO concentration in sample 6 to 6.5 ppm)


**Fig. 10** $NO_2$ measurement with AQUASOL AE257 test kit and $NO_3$ measurement with AQUASOL AE308 test kit

### 5.6 $NO_3$ measurement with AQUASOL AE308 test kit

The procedure is described in part 34 of IS: 3025 [36], and in standard methods for chemical analysis of water and wastewater (method 4500-$NO_3$) [34–36]. Procedure is shown in Fig. 10.

## 6 Results and discussions

Plasma water treatment can be improved by increasing the contact area between water and plasma discharge [14, 15]. The concentrations of the reactive species and the input energy provided decides the effectiveness of the plasma treatment [16]. In the present study ROS and RNS concentrations in water are measured as per international standards [34–36]. Fig. 11 shows the effect of the applied voltage on ROS concentrations in water. Increasing the input voltage has a positive impact on ROS concentrations ($H_2O_2$) till 20 kV, after which the concentration starts to drop down significantly. This shows that output of the plasma treatment process depends not only on the applied voltage but also on the design parameters of the plasma generators such as the volumetric ratio of water and air in the reaction chamber, the treatment time, the gap between the electrode and the water surface, etc. Fig. 11 shows the concentration of $H_2O_2$ in a 1800 mL six compartment plasma generator with in 20 min treatment time. The concentration of the hydrogen peroxide was measured using Aquasol testing kits. The proposed plasma generator design found to be more efficient at 20 kV power, as the highest concentration of $H_2O_2$ produced is 60 ppm. Recent studies on the acidity of the plasma treated water reveal how the acidity affects the organic contaminants removal in water [17]. The acidity in the treated water was due to the reactive nitrogen species generated by the plasma treatment. Fig. 12 shows the effect of the treatment time on pH when 24 kV power is supplied in an 1800 mL six compartment plasma generator. The treatment time varied from 2 to 20 min. The water is the most

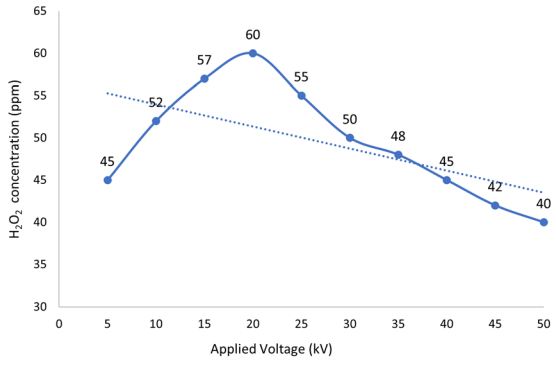


**Fig. 11** Effect of applied voltage on ROS production. Concentration of hydrogen peroxide in 1800 mL MCDBD plasma generator in 20 min treatment time. Dotted line indicates trend of $H_2O_2$ drop with respect to applied voltage.

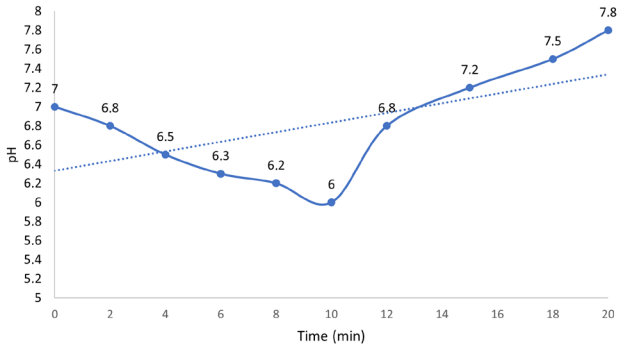


**Fig. 12** Effect of treatment time on water acidity (pH), power supply 24 kV, 1800 mL MCDBD plasma generator. Dotted line shows changes in pH with respect to treatment time.

acidic after 10 min, as pH is found to be 6. The sample was treated for different duration. After 10 min the pH started to increase and became basic in 20 min. The decrease in pH until the 10-min mark is attributed to the fact that acidic species (RNS) are formed in the samples due to the plasma discharge. After this phase the ROS production is significant due to this neutralization and the purification of water is observed. The research done by Lukes et al. [18] concisely shows that ·OH radicals are the key components of any treatment using plasma discharge related processes. This is done by either the removal of hydrogen atoms or electrophilic addition of the double bonding compounds or electron migration. In case of $H_2O_2$ reactions, when metallic ions are considered to specifically generate ·OH radicals from decomposition of $H_2O_2$ then the contaminant degradation efficiency increased. In case of ·NO reactions, the main effect produced is the change of the acidity of the treated water [19]. The production of ROS and RNS in single and six plasma jet reactors are compared in Fig. 13 and Fig. 14. The power supplied was 12 kV – 60 kV and the quantity of water taken was 1000 mL – 1800 mL (166 mL to 300 mL / compartment) for a six compartment plasma generator or MCDBD reactor. The power supplied was 12 kV – 60 kV and the quantity of water taken was 150 mL – 500 mL for a single compartment plasma generator (conventional design). The highest concentration of ROS in 1800 mL water is found to be 0.93 M at 18 min for the six compartment MCDBD type plasma generator. Whereas the highest concentration of ROS was 0.6 M in 500 mL water at 12 min for the single reaction chamber type conventional plasma generators. While in case of RNS the maximum concentration was 0.52 M in the six compartment type plasma generators at 16 min in 1800 mL water. The RNS maximum concentration was found to be 0.44 M for single compartment type plasma generators at 12 min in 500 mL water. Thus, higher concentration of ROS and RNS can be produced and large quantities of water can be treated using MCDBD type plasma generators. The increase in PAW temperature during plasma treatment reduces the life of the active ROS and RNS. Fig. 15 shows the increase in PAW temperature by 1.8 °C in MCDBD, whereas 3 °C in conventional design. It means that the active ROS and RNS

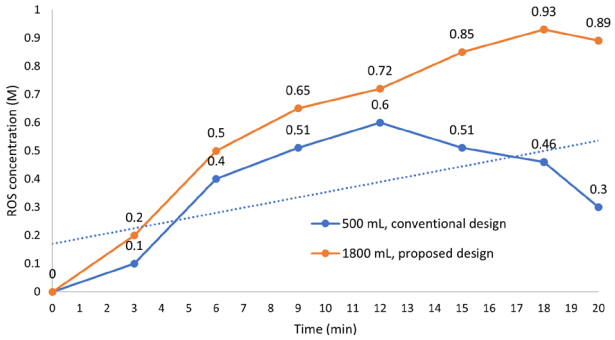


**Fig. 13** ROS production: power supply 24 kV – 1800 mL MCDBD plasma generator (proposed design MCDBD), 24 kV – 500 mL single compartment plasma generator (conventional design). Improvement in ROS concentration represented by dotted line.

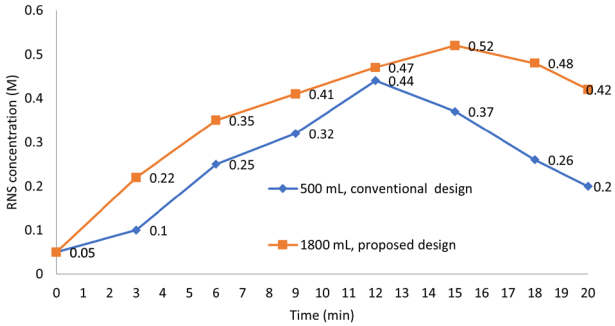


**Fig. 14** RNS production: power supply 24 kV – 1800 mL MCDBD plasma generator (proposed design MCDBD), 24 kV – 500 mL single compartment plasma generator (conventional design)

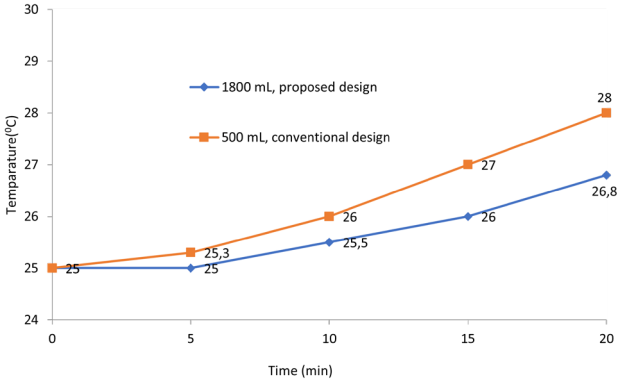


**Fig. 15** Increase in PAW temperature during treatment: power supply 24 kV – 1800 mL six compartment plasma generator (proposed design MCDBD), 24 kV – 500 mL single compartment plasma generator (conventional design)

have longer life in MCDBD reactor which is desirable to deactivate the biofilms and water decontamination.

Even though the same power is supplied to single and multi-compartment plasma generators, the output in terms of concentration of ROS and RNS is found significantly greater in the new designs. The proposed design is more suitable for waste water treatments, biomedical and agricultural applications.

## 7 Conclusions

A novel multi-compartment DBD plasma reactor or generator is developed and tested for water treatment to produce plasma activated water. The efficiency of the plasma reactors greatly depends on the reactive oxygen and nitrogen species produced in PAW. So, in this study the concentrations of ROS and RNS in plasma activated water are evaluated by testing as per international standards [34–36]. Result shows that the output of the plasma treatment process depends not only on high voltage power supply but also on design parameters such as the volumetric ratio of water and air in the reaction chamber, the treatment time, the distance between electrode and water surface. Acidic nitrogen species and efficient oxidizing species formed in the water due to plasma discharge. The highest concentration of ROS in 1800 mL water is found to be 0.93 M at 18 min in an MCDBD type plasma generator. Whereas, the highest concentration of ROS 0.6 M in 500 mL water at 12 min for single jet or conventional plasma generator. The RNS maximum concentration was 0.52 M in MCDBD plasma generator at 16 min of treatment time in 1800 mL water. The RNS maximum concentration was found to be 0.44 M for a single jet plasma generator at 12 min in 500 mL water. Thus, the result shows that higher concentration of ROS and RNS can be produced in higher quantities of water using the proposed MCDBD type plasma generators. The increase in the PAW temperature was 1.8 °C in MCDBD, whereas 3 °C in conventional design. It means that the active ROS and RNS have longer life in MCDBD reactor which is desirable to deactivate the biofilms and water decontamination. The reactive oxygen species produced in PAW are found to be significant for purification and decontamination of water. The proposed design is more suitable for wastewater treatments, biomedical and agriculture applications.

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