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Studying the Properties of Water Activated by Hybrid Plasma for Biological and Medical Applications.

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Abstract

This work investigates the chemical and physical properties of water activated by Hybrid plasma, using argon gas. Two ways were used to activate the water: the first one was done by mixing the argon gas and air at atmospheric pressure at two flow rates of 0.7 and 1.0 L/min, and the second, used only atmospheric air. To study the physical and chemical properties of water, 10 cm^3 of distilled water was placed in a container made of glass in the form of a dish with a diameter of 5 cm and 1 cm depth; an AC sinewave voltage of 12 kV, a frequency of 20 kHz, and power of 36 watts was used; the exposure time ranged from 1-30 minutes. The generated RONS (oxygen nitrogen reactive species) were quantified utilizing tools designed specifically for this objective. The results showed that NO_2, NO_3 and H_2O_2 concentrations increased with the exposure time but they decreased with the increases the flow rate. The water pH in both methods decreased with the exposure time until it reached 2.3; the water temperature increased reaching 51.7°C with the first method and 50.6°C with the second method. The pH increased with the storage time, and the plasma activated water (PAW) reached the natural pH value of 7 after 24 hours of storage. From these findings, it can be concluded that Hybrid plasma discharge can produce RONS that may be applied in biological applications.

Keywords: Hybrid plasma, Oxygen nitrogen reactive species (RONS), plasma-activated water, Continuous corona.

دراسة الخصائص الفيزيائية والكيميائية للمياه التي تنشطها البلازما الهجينة للتطبيقات البيولوجية والطبية.

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الخلاصة

يهدف هذا العمل إلى دراسة الخواص الكيميائية والفيزيائية للماء المنشط بالبلازما الهجينة. تم في هذا العمل دراسة تنشيط الماء بواسطة البلازما الهجينة باستخدام غازالارجون.
تم استخدام طريقتين: الأولى خلط غاز الأرجون مع الهواء بمعدل تدفق 0.7 و 1.0 لتر/دقيقة، والثانية استعمل الهواء الجوي فقط .لدراسة الخواص الفيزيائية والكيميائية للماء، تم وضع 10 سم 3 من الماء المقطر

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في وعاء مصنوع من الزجاج على شكل طبق بقطر 5 سم وعمق 1 سم وتم تنفيذ النظام تحت جهد تيار متناوب 12 كيلو فولت وتردد 20 كيلو هرتز، وبقدره 36 واط، تراوح زمن التعريض من 1−30 دقيقة. تم خلط غاز الأرجون مع الهواء الجوي بمعدل جريان 0.7 و1.0 لتر/دقيقة. تم قياس أنواع RONS (الأنواع التفاعلية للأكسجين والنيتروجين) باستخدام اشرطه مصممة خصيصًا لهذا الغرض. أظهرت النتائج أن تراكيز NO_2 وNO_3 و H_2O_2 تزداد مع الزمن كما تتناقص مع معدل الجريان. كما أن الرقم الهيدروجيني للماء في الطريقة الأولى والثانية يتناقص مع زمن التعرض حتى يصل إلى 2.3 وترتفع درجة الحرارة حتى تصل 51.7 درجة مئوية مع الطريقة الأولى و 50.6درجة مئوية مع الطريقة الثانية ولكن الرقم الهيدروجيني يزداد مع زمن التخزين ويصل الماء إلى حالته الطبيعية مع الرقم الهيدروجيني 7 بعد 24 ساعة من التخزين. من هذه النتائج، يمكن أن نستنتج أن البلازما الهجينه قادره على إنتاج RONS التي يمكن استعملها في التطبيقات البيولوجية.

1-Introduction

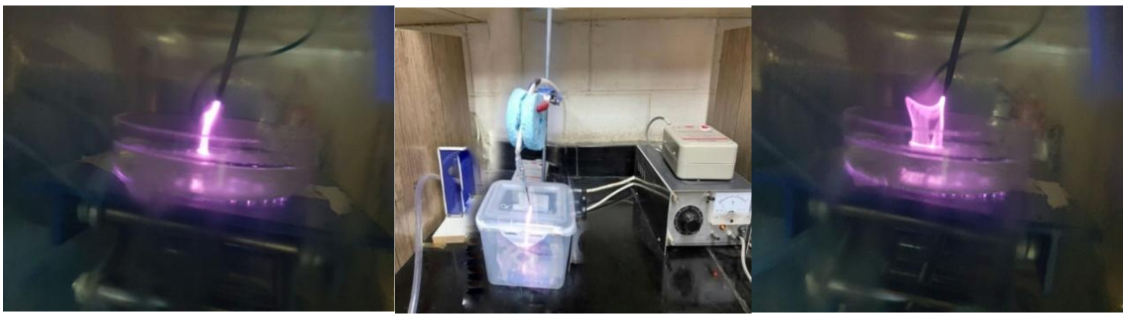
Plasma is an ionized gas that consists of charged particles (electrons, ions, molecules) and is termed 'ionized' due to the presence of one or more free electrons [1, 2]. Plasma-Activated Water (PAW) is produced by plasma water treatment with plasma either above or below the water's surface. This treatment creates various reactive species, particularly reactive oxygen species (ROS) and reactive nitrogen species (RNS). PAW can be used in many applications due to these reactive species, such as agriculture, food industry and healthcare. PAW has also been extensively researched as a novel and flexible antimicrobial because of its reactive species. It is efficient against a wide variety of bacteria [3]. A corona discharge is another type of non-equilibrium weakly ionized plasma. It is a self-sustained electrical discharge that occurs when the surface electric field exceeds the critical starting value, and air is ionized to create plasma; there is no complete breakdown of the gap between the electrodes [4-7]. Corona discharge is separated into two different categories: continuous and pulsed. Continuous corona discharges occur at DC or low-frequency AC voltages. A recent example of work on DC-excited corona discharges is that of Eichwald et al. [8]. A pulsed corona is produced by applying a short (usually sub-microsecond) voltage pulse to an electrode. Its practical benefits lie in the fact that the brief pulse duration prevents the transition to a spark, allowing for operation at higher voltages and currents compared to the continuous corona [9]. Corona discharge are appealing because of their various advantages, including the lack of moving parts, small size, lightweight, and ease of operation. Though other gas discharges, such as dielectric barrier discharges, can produce ionic winds, corona discharges have the benefit of being simple to operate in direct current (DC) mode at atmospheric pressure, resulting in a continuous current [10-13]. Corona discharge finds applications in environments characterized by low concentrations of charged species, such as electrostatic precipitators employed to capture dust particles in numerous industrial settings. Corona discharges are also used to detect radiation and treat the surfaces of polymers [14]. Gao et al. studied the fundamentals of plasma generation, physicochemical properties, and characterization of PAW and the optimization of cold plasma activation by micro bubbles [15]. Herianto et al. studied the PAW applications for enhanced food safety (both biological and chemical safeties). The subsequent effects on food quality (chemical, physical, and sensory properties) are discussed in detail [16]. Tamara and Hammad studied the effect of plasma activated water on escherichia coli bacteria to disinfect the tooth root canal [17]. Sura and Hammad developed low temperature non-thermal atmospheric plasma jet sources for the biological applications [18]. Farah and Hammad studied the effect of gas flow rate, exposure times and ageing on the physicochemical properties of water activated by glow discharge plasma jet [19]. In other studied for the same authors, they studied the physicochemical properties of water activated by microwave-induced plasma jet for biological and medical applications [20]. Guragain et al.

studied the germination and growth of sprouts irrigated using plasma activated water (PAW) [21]. Guragain et al. studied the impact of PAW on seed germination of soybean [22]. Guragain et al. studied the influence of PAW on the germination of radish, fenugreek, and pea seeds [23]. Tamara and Hammad studied the physicochemical properties of water activated by cold atmospheric plasma jet [24]. Tamara and Hammad studied the effect of the water activated by plasma and the direct plasma on enterococcus faecalis bacteria for disinfection of tooth root canal [25].

This research aims to study the chemical and physical properties of water activated with hybrid plasma (corona plasma working with air mixed with argon gas and high frequency power supply) developed for this purpose in order to obtain a plasma more suitable for activating water than other types of plasma used for the same purpose.

2- Experimental works

PAW was generated when water was irradiated with atmospheric pressure argon-air plasma and atmospheric pressure air plasma (corona discharge). The system for producing plasma consists of a power supply capable of supplying a high alternating voltage of up to 12 kV in the form of a sinusoidal wave with a frequency of 20 kHz. The high voltage is connected to a tapered electrode of 10 cm in length. The second electrode is made of aluminum in the form of a plate and placed under a bowl of water in the form of a saucer with a diameter of 5 cm and a depth of 1 cm. Two parameters concerning the irradiation with both plasmas were changed: exposure time of plasma onto the water in the range of 1-30 minutes and the argon gas-air mixture flow rate of 0.7 and 1.0 L/min. The concentrations of the generated NO_2, NO_3, and H_2O_2 species were measured as a function of exposure time and gas flow rate using special kits for this purpose. The acidity and temperature of the water were also measured. The acidity was measured with a pH meter; and the temperature with an IR thermometer. The concentrations of NO_2, NO_3, H_2O_2 and acidity were also measured after ending the exposure to plasma and for different periods of time to determine the effect of storage time on the chemical and physical properties of the activated water. Figure 1 shows the plasma system used for water activation.



A (Atmospheric air only) **B** (diagram of hybrid corona) **C** (mixing Argon gas with atmospheric air)

Figure 1: Image for the hybrid plasma used for water activation at working

3-Results and discussion

Table 1 presents the concentrations of NO_2, NO_3, H_2O_2 and the total concentrations as a function of the the exposure time and gas flow rates of 0.7 and 1.0 L/min. for atmospheric pressure argon-air plasma. Table 2 shows the concentrations of NO_2, NO_3, H_2O_2 and the total concentrations as a function of the exposure time for corona discharge irradiation.

Figure 2 shows the relationship between NO_2, NO_3, and H_2O_2 concentrations (in ppm units) and the exposure time (in minutes) at gas flow rates 0.7 and 1.0 L/min. for atmospheric pressure argon-air plasma. Figure **3** shows the relationship between NO_2, NO_3, and H_2O_2 concentrations (in ppm units) and the exposure time (in minutes) for corona discharge irradiation. The concentrations of NO_2, NO_3, and H_2O_2 increased with increasing the exposure time, reaching their highest value at slightly different exposure times. This difference is due to the difference in the half-life of these reactive species. Another factor reinforced this difference, which is the oxidation of NO_2 to NO_3.

From Table 1 and 2, Figures 2 (A, B) and Figure 3, it is evident that the primary factor influencing the concentrations of the reactive species (NO_2, NO_3, and H_2O_2) is the exposure time. Longer exposure times result in higher concentrations of reactive species. Specifically, when mixing argon gas with atmospheric air at flow rates of 0.7 and 1.0 L/min. The optimal exposure time to produce the highest reactive species was found to be 20 minutes. While when utilizing corona plasma (air only), the optimal exposure time was 30 minutes. The second influencing factor is the flow rate. The optimal flow rate was 0.7 L/min (this flow rate represents flow without swirls), but at flow rates greater than 1 L/min, the gas flow speed becomes greater than that at lower flow rates, which leads to the displacement of the air, which consists mainly of oxygen and nitrogen. The air is replaced by argon gas, which has been added to the system in a high percentage. Oxygen and nitrogen in air are necessary in the formation of the reactive species. The third factor is the different half-lives of NO_2, NO_3, and H_2O_2. The fourth factor is the possibility of converting NO_2 into NO_3 through a series of chemical reactions maintained by the plasma.

Table 1: (A) The concentrations of NO_2, NO_3, H_2O_2 and the total concentrations as a function of the exposure time at 0.7 L/min gas flow rate for atmospheric pressure argon gas-air mixture plasma was used for irradiation.

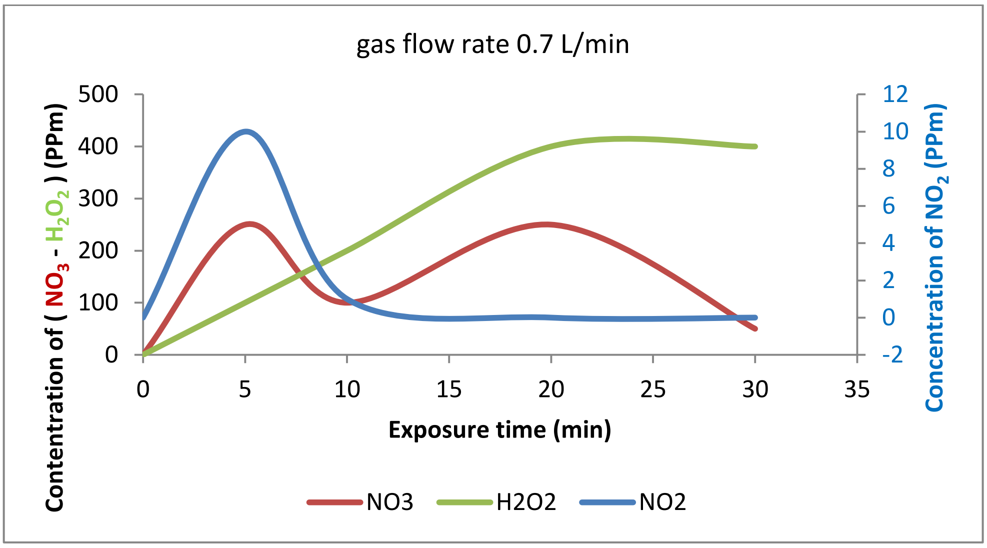
Exposure time(min)	NO_2	NO_3	H_2O_2	Total concentrations
0	0	0	0	0
5	10	250	100	360
10	1	100	200	301
20	0	250	400	650
30	0	50	400	450

(B) The concentrations of NO_2, NO_3, H_2O_2 and the total concentrations as a function of the exposure time at 1.0 L/min gas flow rate for atmospheric pressure argon gas-air mixture plasma was used for irradiation.

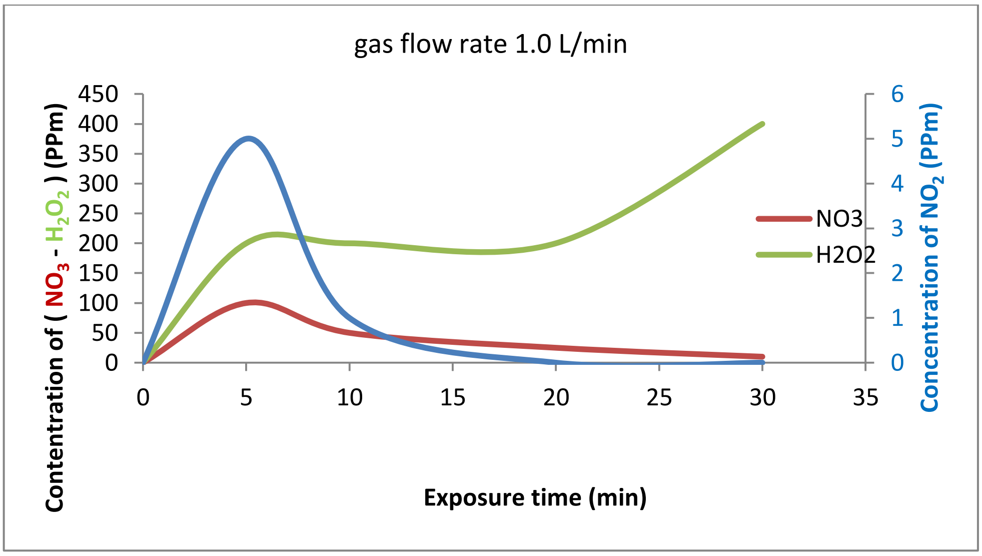
time(min)	NO_2	NO_3	H_2O_2	Total concentrations
0	0	0	0	0
5	5	100	200	305
10	1	50	200	251
20	0	25	200	225
30	0	10	400	410

Table 2: The concentrations of NO_2, NO_3, H_2O_2 and the total concentrations as a function of the the exposurer time for irradiation with corona discharge(only air).

time(min)	NO_2	NO_3	H_2O_2	Total concentrations
0	0	0	0	0
5	10	250	100	360
10	1	250	200	451
20	10	500	0	510
30	25	500	0	525



A



B

Figure 2: The relationship between NO_2, NO_3, and H_2O_2 concentrations (in ppm) and the exposure time (in minutes) for atmospheric pressure argon-air plasma irradiation at (A) 0.7 L/min and (B)1.0 L/min flow rates.

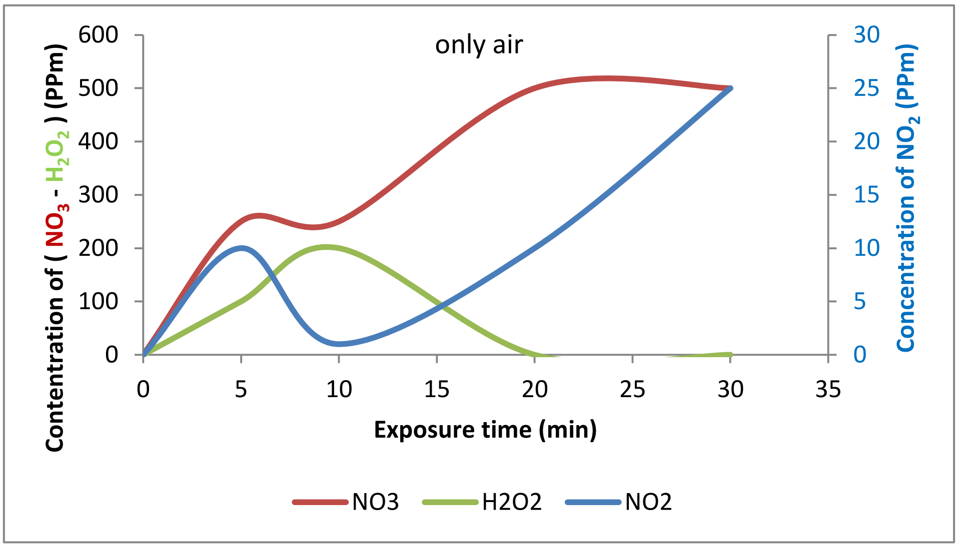


Figure 3: The relationship between the concentrations of NO_2, NO_3, and H_2O_2 and exposure time for irradiation with corona discharge (only air).

Figure 4 shows the relationship between exposure time and pH values of PAW for irradiation with atmospheric-pressure argon gas-air mixture plasma at two gas-air mixture flow rates of 0.7 and 1.0 L/min. Figure 5 shows the same relationship in the case of air only (irradiation with corona discharge). In both cases, a noticeable decrease in pH occurred; pH decreased from 7 to 2.3 within 30 minutes when water was exposed to argon-air mixture, irrespective of the flow rate. Similarly, a pH decrease from 7 to 2.4 within 30 minutes was noted for irradiation with corona discharge, as seen in Fig. 5. This pH reduction can be attributed to the formation of compounds due to the irradiation of water with plasma, resulting in increased water acidity. Nitric acid is one such compound formed through the interaction of NO_3 with hydrogen produced during water dissociation through plasma irradiation. These acidic solutions generated by the treatment process are highly effective in reducing the activity of pathogens.

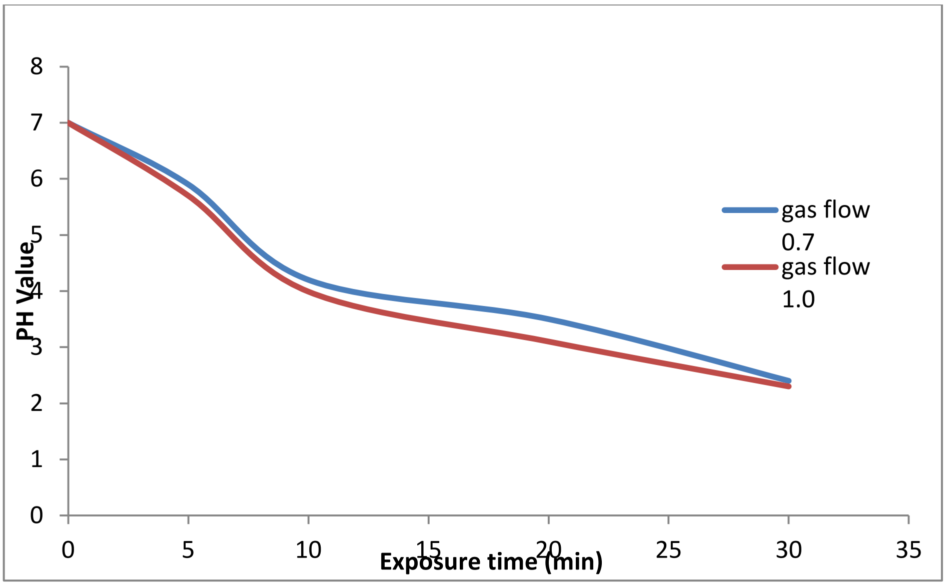


Figure 4: pH change with exposure time for atmospheric-pressure argon-air plasma irradiation at flow rates of 0.7 and 1.0 L/min.

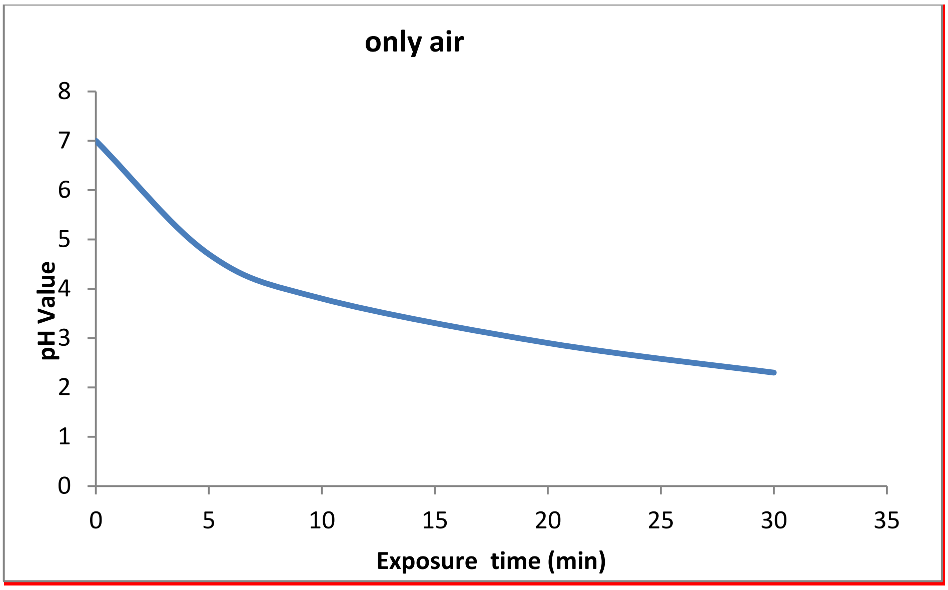


Figure 5: The relationship between pH and exposure time for corona discharge irradiation (only air).

Figures 6 and 7 illustrate the correlation between the irradiated water temperature and exposure time. Figure 6 shows this relation for water irradiatd with atmospheric-pressure argon-air plasma at two flow rates, 0.7 and 1.0 L/min. Meanwhile, Figure 7 portrays the results of corona discharge irradiation. It is evident from these figures that the water temperature increased as the exposure time increased. The highest water temperature of approximately 51.7^{o} Celsius was recorded during the irradiation to atmospheric-pressure argon-air plasma at two argon-air mixture flow rates, 0.7 and 1.0 L/min. In the case of corona discharge irradiation, the highest temperature was 50.6^{o} Celsius.

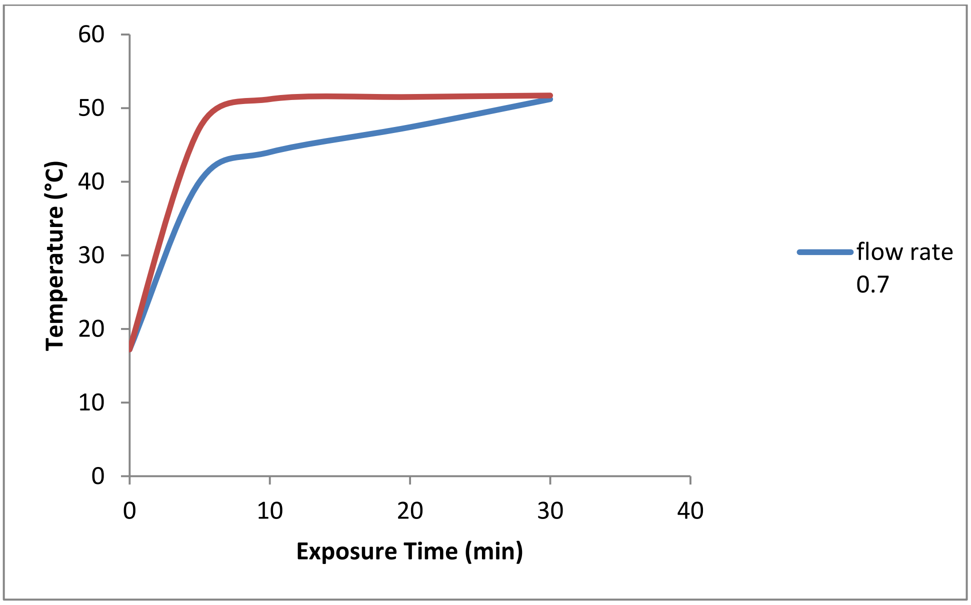


Figure 6: Water temperature as a function of exposure time for atmospheric-pressure argon-air plasma irradiation at at two argon-air mixture flow rates of 0.7 and 1.0 L/min.

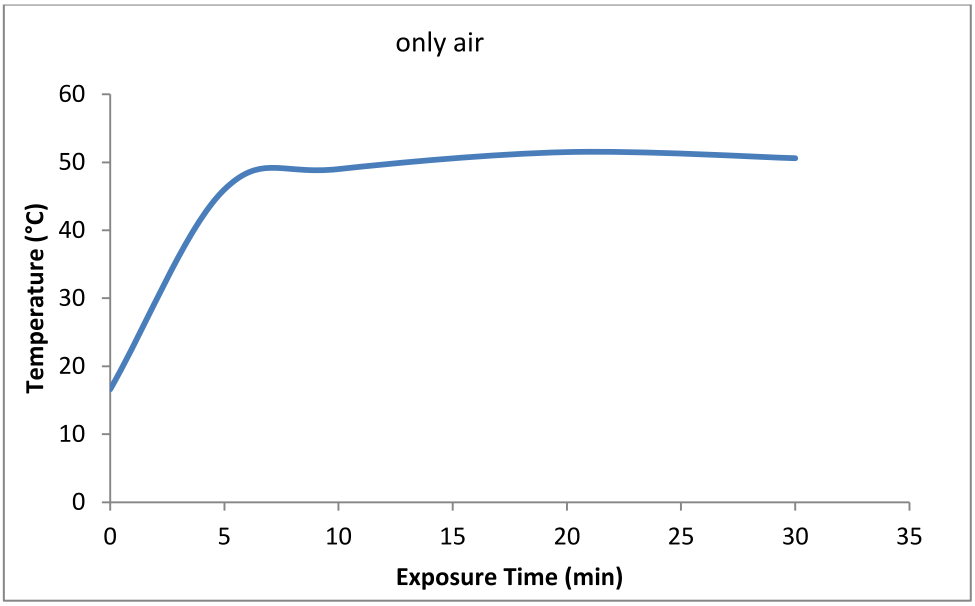


Figure 7: Water temperature as a function of exposure time for corona discharge irradiation (the case of air only).

Figure 8 displays the correlation between water storage time and the concentrations of NO_2, NO_3, and H_2O_2 concentrations (in ppm) for argon-air plasma irradiation at 0.7 and 1.0 L/min argon-air mixture flow rates. Figure 9 illustrates the same relation for for corona discharge irradiation (the case of air only). Figures 8(A, B) and 9 show that the concentrations of NO_2, NO_3, and H_2O_2 gradually declined over time. Notably, for argon-air plasma irradiation, these reactive species reached half of their initial concentrations after 6-hour of storage at room temperature. For plasma not mixed with argon gas, the reduction to half of its initial concentrations occurs within the same time of 6 hours, as detailed in Table (3,4). It was also noted from Table (3) that the concentration of NO_2 reached zero within a short time. This phenomenon is not clear, it requires further study. Furthermore, it is noteworthy that all reactive species concentrations reached low values after 30 hours of storage. This pattern is contingent upon the respective half-life of these reactive species.

Table 3: A Concentrations of NO_2, NO_3, H_2O_2 and the total concentrations as a function of storage time for argon-air plasma irradiation at (A) 0.7 L/min gas flow rate, (B) 1.0 L/min gas flow rate

A

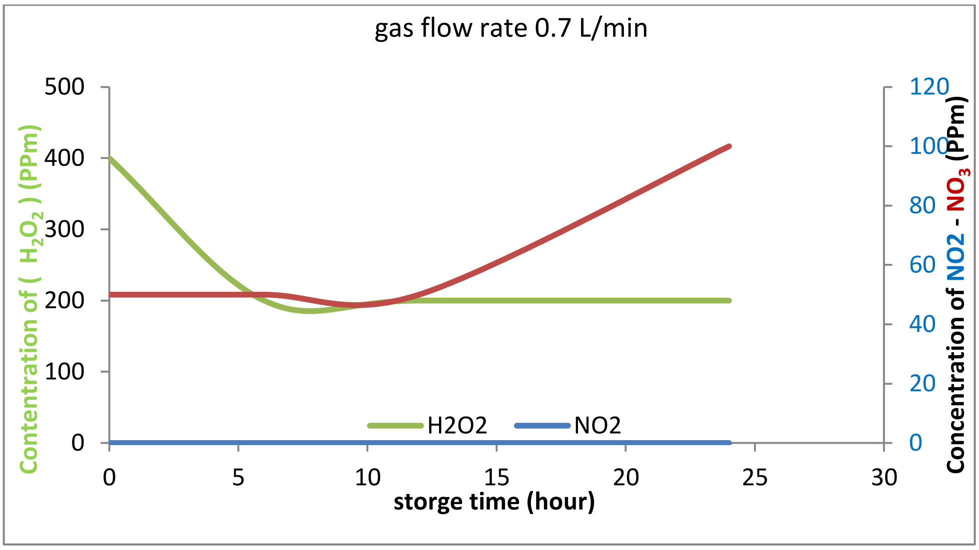
Time (hour)	NO_2	NO_3	H_2O_2	Total concentrations
0	0	50	400	450
6	0	50	200	250
12	0	100	200	300
24	0	50	200	250

B

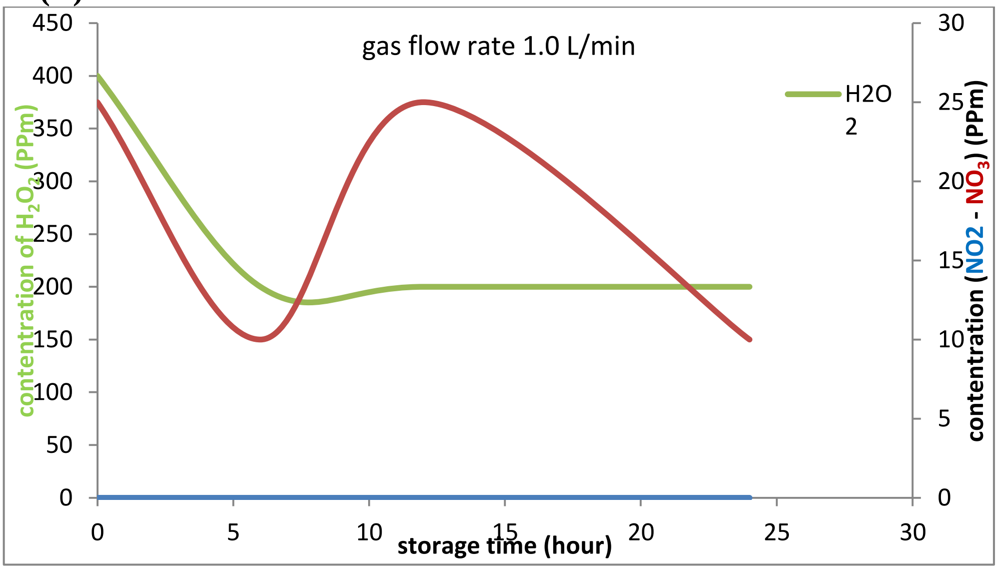
Time (hour)	NO_2	NO_3	H_2O_2	Total concentrations
0	0	25	400	425
6	0	10	200	210
12	0	25	200	225
24	0	10	200	210

Table 4: Concentrations of NO_2, NO_3, and H_2O_2 and the total concentrations as a function of storage time for corona discharge irradiation (only air).

TIME (HOUR)	NO_2	NO_3	H_2O_2	total concentrations
0	25	500	0	525
6	10	500	0	510
12	0.5	500	0	500.5
24	0	500	0	500



(A)



(B)

Figure 8: The correlation between water storage time and the concentrations of NO_2, NO_3, and H_2O_2 (in ppm) for argon-air plasma at (A)0.7 L/min gas flow rate, (B)1.0 L/min

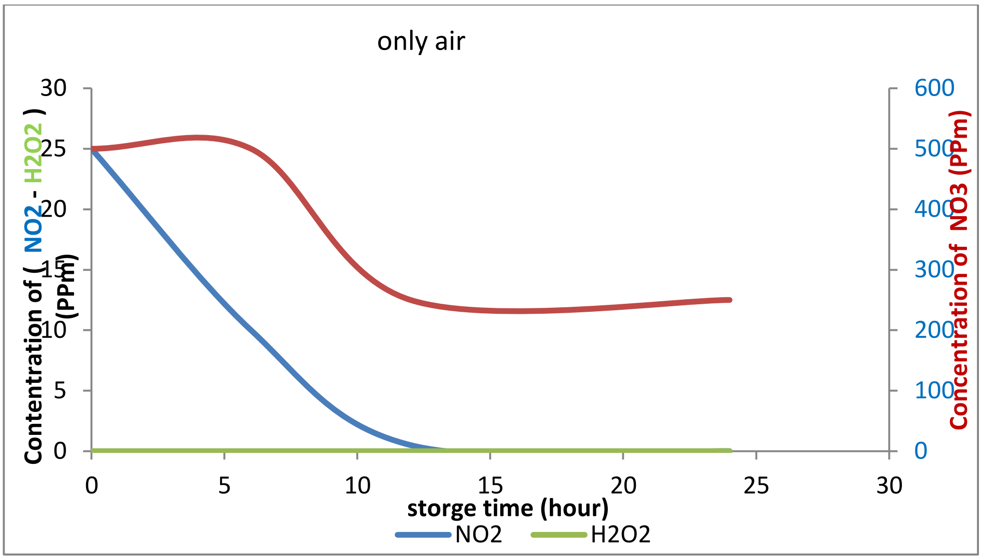


Figure 9: The relationship between the concentration of NO_2, NO_3 and H_2O_2 of the water irradiated with corona discharge and water storage time.

Figures 10 and 11 illustrate the correlation between the storage duration of PAW and its pH for the plasma that works with a mixture of air and argon gas and the other case with only air. The data from these figures clearly indicates that as the storage time increases, the pH of the activated water rises, eventually stabilizing at a neutral pH of 7 after 24 hours of storage. This pH adjustment occurs because the reactive species (NO_2, NO_3 and H_2O_2) within the water gradually transform into more stable components, ultimately restoring the water to its natural state.

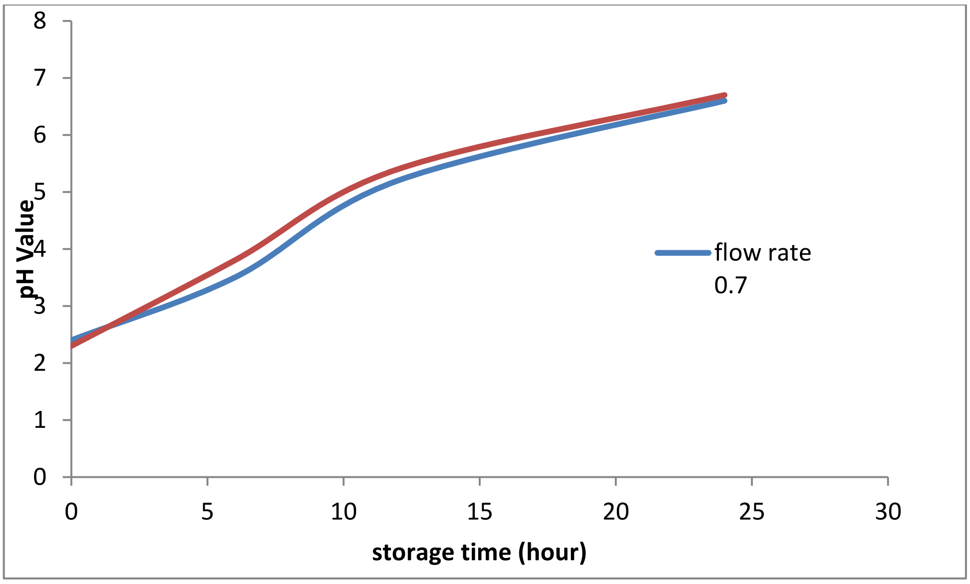


Figure 10: pH water changes with storage time for argon-air plasma at flow rates of 0.7 and 1.0 L/min.

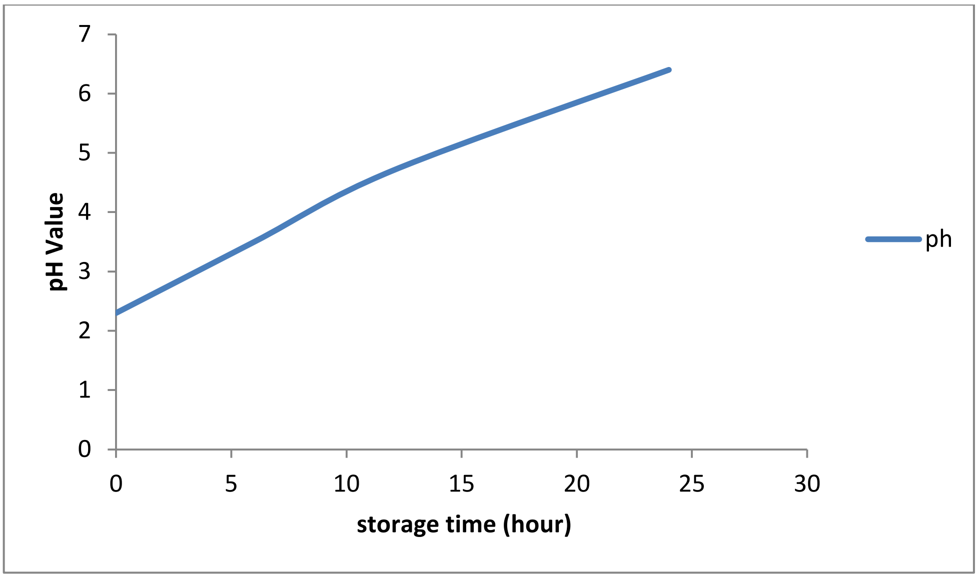


Figure 11: pH water changes with storage time for the plasma that is not mixed with argon gas.

4- Conclusions

This research explores the potential for water activation through Hybrid plasma, resulting in the generation of Reactive Oxygen and Nitrogen Species (RONS). When water was irradiated with atmospheric-pressure argon gas-air plasma at flow rates of 0.7 and 1.0 L/min, the concentration of these species steadily increased with longer exposure times. Longer exposure durations led to higher concentrations. The optimal exposure time of atmospheric-pressure argon gas-air plasma was 20 minutes for a flow rate of 0.7L/min, while it was 30 minutes for corona discharge irradiation. As for the effect of the flow rate of the gas-air mixture in the atmospheric-pressure argon gas-air plasma irradiation, flow rate of 0.7L/min produced higher concentrations of the reactive species than the 1.0L/min. Another factor to consider is the different half-lives of NO_2, NO_3, and H_2O_2. Among these species, H_2O_2 has the longest half-life, followed by NO_3, with NO_2 having the shortest. Additionally, the conversion of NO_2 into NO_3 through a series of chemical reactions facilitated by plasma also has an effect also. Furthermore, the presence of argon gas results in a slightly higher water temperature (51.7 °C) than when there was no argon gas (50.6 °C). The water pH decreased with exposure time of atmospheric-pressure argon gas-air plasma and corona discharge irradiations, but after 24 hours of storage, it returned to its initial value. In conclusion, the activation of water through Hybrid plasma is a straightforward and eco-friendly method with potential applications in the medical and biological fields.

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