

THE EFFECT OF GAS FLOW RATE, EXPOSURE TIMES AND AGEING ON THE PHYSICO-CHEMICAL PROPERTIES OF WATER ACTIVATED BY GLOW DISCHARGE PLASMA JET[†]

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The goal of this work is to look into how the glow discharge plasma jet system changes the chemical and physical features of water. In this work, the physical and chemical properties of water were studied by using a plasma jet with Argon gas. 10 cm³ of distilled water was put in a glass dish with a diameter of 5 cm and a depth of 1 cm. The system was run with an AC voltage of 12 kV and a frequency of 20 kHz, and the exposure time ranged from 1 to 30 minutes. With amounts of 0.7, 1.0, 1.5, and 2.1 l/min, kits made by the American company Bartvation were used to measure the types of reactive oxygen and nitrogen species (RONS) that were formed. The data showed that the levels of NO₂, NO₃, and H₂O₂ were all too high. It gets bigger over time and as the flow rate goes up. The pH goes down with time until it hits 3, and the temperature goes up until it reaches 33°C. However, the pH goes up with storage time, and after 24 hours the water is back to its natural pH of 7. The amount of NO₂, NO₃, in the air goes up a little bit, and then starts to go down rapidly after 6 hours. After 24 hours, it is close to zero. From this, it's clear that the glow discharge plasma jet device can make RONS, which can be used for biological purposes.

Keywords: Non-Thermal Plasma (NTP); Direct Current (DC); Plasma Activated Water (PAW)

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1. INTRODUCTION

Non-thermal plasma (NTP) is an ionized gas—the fourth state of matter—that generates reactive chemical species, such as reactive oxygen and nitrogen species, electrons, atoms, neutral molecules, charged species, and ultraviolet radiation [1]. NTP can be artificially generated from ambient air or certain gases at atmospheric and low pressures in the presence of a high voltage (~kV) electric current, current (AC), or direct current (DC) can be used for plasma discharge [2]. There are several types of plasma systems suitable for biological and medical applications. The most widely used plasmas are atmospheric pressure dielectric barrier discharges DBD, plasmas jet, and coronary discharges [3,4,5]. One popular technique has been the application of plasma into water to produce plasma-activated water (PAW). The PAW technology has been used in many applications such as disinfection against bacteria, as fertilizer, in chemical reduction, and in water treatment disinfection and sterilization applications, dermatology, cancer treatment, dentistry, anti-fungal treatment, and wound healing. PAW is considered a technology that has various applications, and it is an ecofriendly and cost-effective disinfectant [6]. The key inactivation agents of PAW are the creation of reactive oxygen species (ROS) (atomic oxygen (O), superoxide (O₂⁻), hydrogen peroxide (H₂O₂), hydroxyl radicals (OH•), and singlet oxygen ¹O₂ [7], these components have a role later in the process of activating the water. Also, reactive nitrogen species RNS such as NO₂, NO₃ and proxy nitrite will be form [8, 9]. When these compounds reach the surface of the liquid, they will dissolve in the liquid (water) and change its chemical and physical properties. In addition, the non-thermal plasma interaction with the liquid leads to the dissolution of molecules and a very effective layer of electrons. The molecules fragment of the liquid and the interaction of the reactive species of oxygen and nitrogen RNOS with the layer of electrons on the surface of the water leads to an increase in the hydrogen number of the water PH and formation of free radicals that will contribute a lot to the applications of PAW when it is used in the medical or biological field [10]. Non-thermal plasma can be used directly in disinfection and sterilization applications, dermatology, cancer treatment, dentistry, anti-bacterial and anti-fungal treatment, and wound healing [11-14]. They studied compare the exposure of the tooth root canal contaminated with Escherichia coli bacteria to plasma jet and plasma-activated water. The obtained results found that the response of bacteria to treatment with plasma jet is better than treatment with activated water with plasma [15]. Surat Abdul-Kadhim and Hamad R. Hammoud studied Building a plasma jet system for biological purposes. The developed plasma system consists of a power source and a plasma torch. The results showed that the developed plasma is suitable for biological applications [16]. I.E. Vlad et al found that PAW treatment caused a significant reduction in the viability of all bacterial strains tested. The bacterial inhibition effect was found to be dose-dependent, with higher doses resulting in greater inhibition. The authors also investigated the mechanism of action of PAW and found that it induced oxidative stress and DNA damage in the bacteria, leading to their death [17]. Yinglong Li et al utilized a plasma jet device to generate PAW and tested its antimicrobial effect against three strains of bacteria: Streptococcus mutans, Porphyromonas gingivalis, and Candida albicans. The study found that PAW significantly reduced the viability of all three strains of bacteria in a dose-dependent manner [18]. Renwu Zhou et al studied different methods used to generate PAW and the effects of plasma parameters on the generation of reactive species. The authors also discuss the

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mechanisms of reactive species formation in PAW and their potential biological applications, including disinfection, wound healing, and cancer treatment [19]. Tomy Abuzairi et al studied plasma treatment using atmospheric pressure plasma was demonstrated. Physicochemical properties such as pH, temperature, ORP, and nitrite concentration were assessed. The results suggest that plasma treatment causes acidification on water and generate reactive species, creating an environment suitable for killing bacteria [20]. Rajesh Prakash Guragain et al studied PAW is tested on four seed species: phapar (*Fagopyrum esculentum*), barley (*Hordeum vulgare*), mustard (*Brassica nigra*), and rayo (*Brassica juncea*). The line-frequency air-operated gliding arc discharge (GAD) system treated DI water for 5- or 10-min. PAW had significantly different physicochemical characteristics than DI water. DI water temperature remained unchanged after GAD exposure. PAW improved all calculated germination parameters compared to the control. They also increased seedling length and vigor [21].

This work will work on developing a glow-discharge plasma jet system. Then a certain amount of water activates after placing it in a suitable container with the two types of plasma. We depend on exposure time, gas type, and flow rate as basic variables. Then we are working to determine the amount of NO_2 , NO_3 and H_2O_2 as a function of time and flow rate. The quantitative and qualitative analysis is carried out by means of kits manufactured for this purpose. In addition to PH and water temperature. Then we determine the best conditions to obtain the largest amount of activated water and the highest concentrations of the RONS.

2. EXPERIMENTAL WORKS

The water was irradiated by plasma jet system. Fig. 1 show the system used for the irradiation. The system 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 was connected to an electrode made of aluminum wrapped at the end of a glass tube with inner diameter of 5 mm at a distance of one centimeter from the end of the tube. About 2 cm separates the tube from the water, and the flow is so small that we can't see any swirls on the surface. The second electrode of the system, which was in the form of aluminum plate, was placed under the water container as shown in Fig. 1.

The glass tube at the other end was connected to a bottle of argon gas through a flow rate regulator. 10 cm³ of distilled water was placed in a container made of glass in the form of a dish with a diameter of 5 cm and a depth of 1 cm. This work was done by a group of researchers, some of whom work on microscopes (meaning spectral studies), some on plasma-activated water (meaning bacteria and viruses), and some on creating plasma systems for activating water. For the purpose of studying the effect of plasma on water, we adopted two variables, the first is the exposure time at intervals, where the longest exposure time was 30 minutes, and the second was the flow rate of argon gas, where the flow rate adopted at 0.7, 1.0, 1.5 and 2.1 l/min. After that, the concentrations of NO_2 , NO_3 , and H_2O_2 were measured as a function of exposure time and flow rate of argon gas, using special kits for this purpose. The pH and temperature of the water were also measured. The acidity was measured with a pH scale and temperature using an IR thermometer. The concentrations of NO_2 , NO_3 , H_2O_2 and acidity were also measured after stopping exposure to plasma and for different periods of time in order to determine the effect of storage on the chemical and physical properties of activated water.

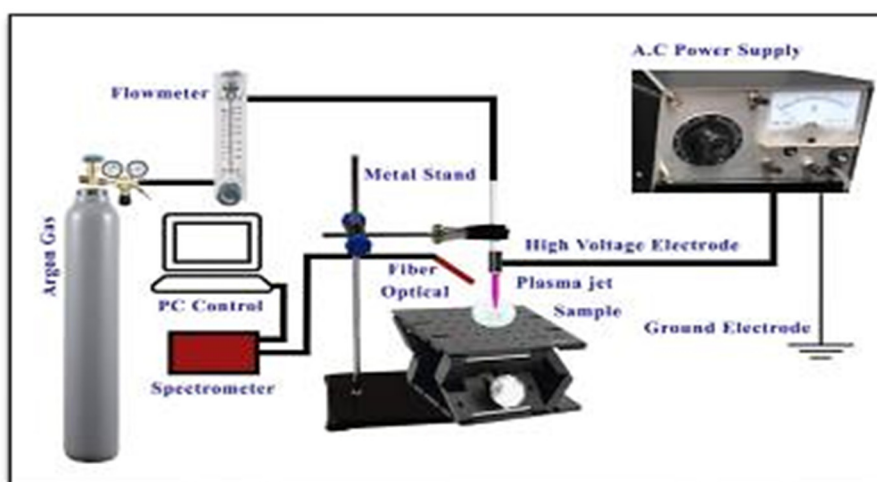


Figure 1. Plasma Jet system at working

3. RESULTS AND DISCUSSION

Fig. 2 shows the relationship between the concentration of NO_2 , NO_3 , H_2O_2 in units ppm and the exposure time at argon gas flow rate, A – for gas flow rate 0.7 l/min, B – gas flow rate of 1.0 l/min, C – gas flow rate of 1.5 l/min and D – gas flow rate of 2.1 l/min. From Fig. 2 (A, B, C, and D,) we notice that the concentrations of the active species increase dramatically (up and down) with the exposure time.

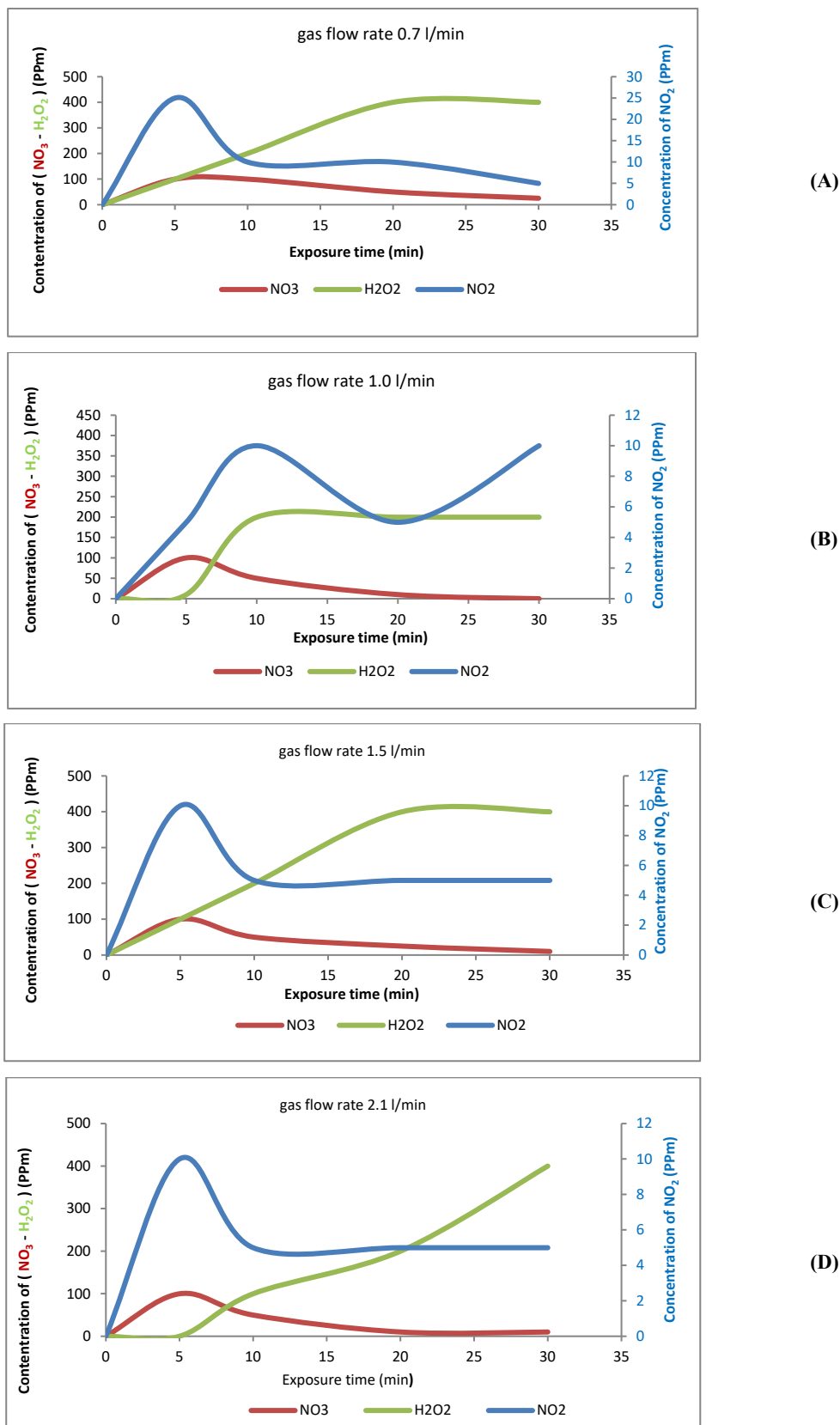


Figure 2. The relationship between the concentration of NO₂, NO₃, H₂O₂ in unit's ppm and the exposer time at argon gas flow rate: A – for gas flow rate 0.7 l/min, B – gas flow rate of 1.0 l/min, C – gas flow rate of 1.5 l/min, D – gas flow rate of 2.1 l/min.

We notice that each of the concentrations of NO₂, NO₃, and H₂O₂ increases with increasing exposure time, and reaches its highest value at slightly different times. This difference is due to the different in the half-life of these active

species, in addition to that there is conversion between these active species Table 1 shows the total concentrations of NO_2 , NO_3 and H_2O_2 for different gas flow rate. From Figure 2A, 2B, 2C, 2D and Table 1A, 1B, 1C and 1D it is clear that the main factors that control the concentrations of the active elements are the exposure time, as the longer the time, the greater the concentration of the active species, and the best irradiation time is 20 minutes. The second factor is the flow rate. Where the best flow rate is 0.7 l/min where this flow is laminar flow and does not contain swirls. At flow rates greater than 1 L/min, the gas flow velocity becomes greater, and this leads to displacement of atmospheric air located in front of the plasma jet, and thus the percentage of atmospheric gas decreases. The percentage of Ar gas is higher because the system works with Argon gas. The third factor is the different in the half-lives of the NO_2 , NO_3 , H_2O_2 , where the longest-lived is H_2O_2 , followed by NO_3 , and the shortest-lived is NO_2 . The fourth factor is the possibility of converting NO_2 into NO_3 through a series of chemical reactions that are sustained by plasma. From Table 1 we note that the best irradiation time is 20 minutes, as we get at this time the largest amount of active elements. The best flow rate is 0.7 l/min. Where the greater amount of the active species operands 460 ppm, which is the highest quantity for different flow rates. There is currently no method to determine which of these four elements is the most important; nonetheless, we believe that length of exposure time is the most important, followed by the chemical interactions between these concentrations and their transformations.

Table 1. The total concentrations of NO_2 , NO_3 , H_2O_2 for different gas flow rate values

Gas flow rate 0.7 l/min				
TIME (MIN)	NO_2	NO_3	H_2O_2	total concentrations
0	0	0	0	0
5	25	100	100	225
10	10	100	200	310
20	10	50	400	460
30	5	25	400	430
Gas flow rate 1.0 l/min				
TIME (MIN)	NO_2	NO_3	H_2O_2	total concentrations
0	0	0	0	0
5	5	100	10	115
10	10	50	200	260
20	5	10	200	215
30	10	0	200	210
Gas flow rate 1.5 l/min				
TIME (MIN)	NO_2	NO_3	H_2O_2	total concentrations
0	0	0	0	0
5	10	100	100	210
10	5	50	200	255
20	5	25	400	430
30	5	10	400	415
Gas flow rate 2.1 l/min				
TIME (MIN)	NO_2	NO_3	H_2O_2	total concentrations
0	0	0	0	0
5	10	100	0	110
10	5	50	100	155
20	5	10	200	215
30	5	10	400	415

Fig. 3 shows the relationship between the pH as a function of exposure time. At a flow rate rate 0.7 l/min, 1.0 l/min, 1.5 l/min and 2.1 l/min. From the figure, we notice a decrease in the pH from 7 to 3 during 30 minutes of exposure. And the behavior is similar in the four case of flow. The low PH was due to the treatment of water with plasma, where the treatment leads to the formation of compounds that increase the acidity of the water, such as nitric acid, which results from interaction of NO_3 with hydrogen, which results from the disusation of water by the effect of plasma. Acidic solutions have a high effectiveness in reducing the effectiveness of pathogens.

Fig. 4 shows the relationship between water temperature and plasma exposure time at a flow rate 0.7, 1.0, 1.5 and 2.1 l/min. From the figure, it is clear that there is an increase in the temperature of the water, and that the highest temperature the water reached is about 33°C , as this degree is still not severe and close to the room temperature. We also note that the change in temperature rchanges slightly different with the flow rate.

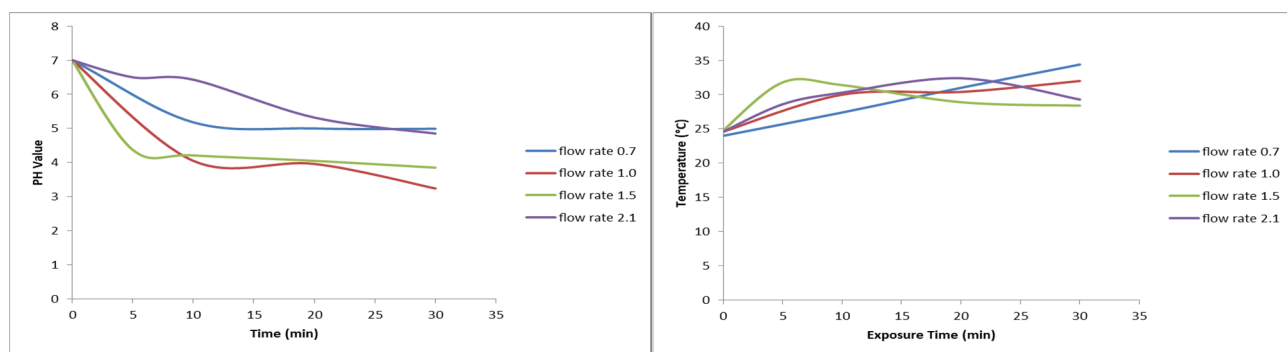


Figure 3. The relationship between the pH and the exposer time at argon gas flow rate 0.7, 1.0, 1.5 and 2.1 l/min

Figure 4. The relationship between the the water temperature and the exposer time at argon gas flow rate of 0.7, 1.0, 1.5 and 2.1 l/min

Figure 5 shows the relationship between the concentration of NO₂, NO₃ and H₂O₂ in units ppm and the water storage time at different argon gas flow rate. A 0.7 l/min, B 1.0 l/min, C 1.5 l/min, D 2.1 l/min. From the Figure 5 A, B, C, and D, we notice that the concentrations of NO₂, NO₃, H₂O₂ decrease with time, and that the concentrations of these reactive species reach half their concentration after 6 hours of storing at room temperature. As shown in Table (2), we also notice that all reactive species concentration reach small values after 24 hours of storage, and this behavior depends on the half-life time of these reactive species.

Table 2. Total concentrations of NO₂, NO₃, H₂O₂ funtion of storage time for different gas flow rate

Gas flow rate 0.7 l/min				
TIME (HOUR)	NO ₂	NO ₃	H ₂ O ₂	total concentrations
0	10	25	200	235
6	5	50	200	255
12	5	50	100	155
24	1	0	100	101
Gas flow rate 1.0 l/min				
TIME (HOUR)	NO ₂	NO ₃	H ₂ O ₂	total concentrations
0	5	10	200	215
6	5	0	200	205
12	1	0	200	201
24	1	0	0	1
Gas flow rate 1.5 l/min				
TIME (HOUR)	NO ₂	NO ₃	H ₂ O ₂	total concentrations
0	5	10	400	415
6	1	0	100	101
12	1	0	0	1
24	1	0	0	1
Gas flow rate 2.1 l/min				
TIME (HOUR)	NO ₂	NO ₃	H ₂ O ₂	total concentrations
0	5	100	0	105
6	1	10	0	11
12	1	0	0	1
24	1	0	0	1

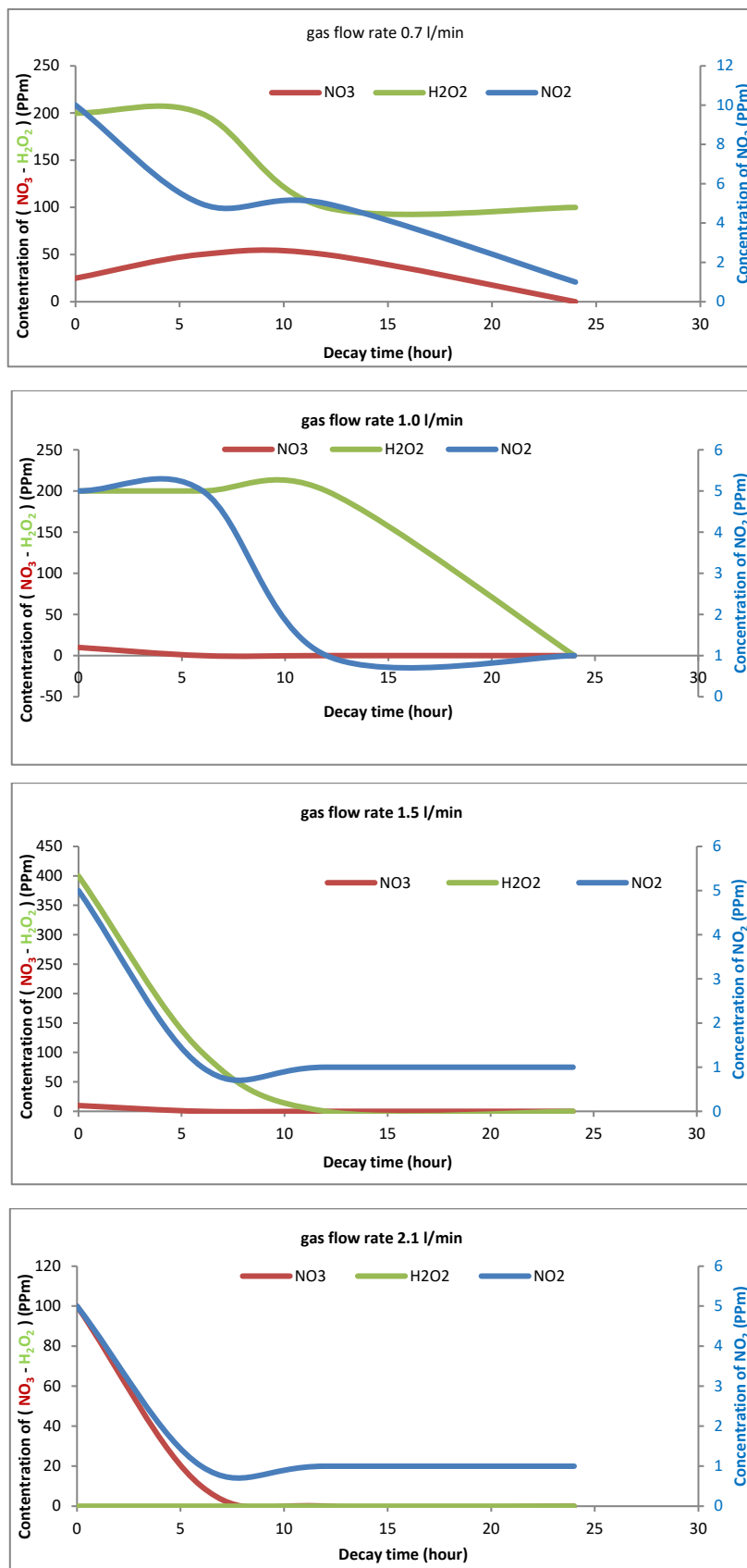


Figure 5. the relationship between the concentration of NO_2 , NO_3 and H_2O_2 in units ppm and the water storage time at different argon gas flow rate. A 0.7 l/min, B 1.0 l/min, C 1.5 l/min, D 2.1 l/min.

Fig. 6 shows the relationship between the storage time of the PAW and the PH of the water activated at a flow rate 0.7,1.0,1.5 and 2.1 l/min. It is clear from the figure that the pH increases with the storage time and the water reaches its

natural pH of 7 after 24 hours of storage this is because the reactive species upon storage have disappeared and turned into more stable elements and thus the water has returned to its normal state.

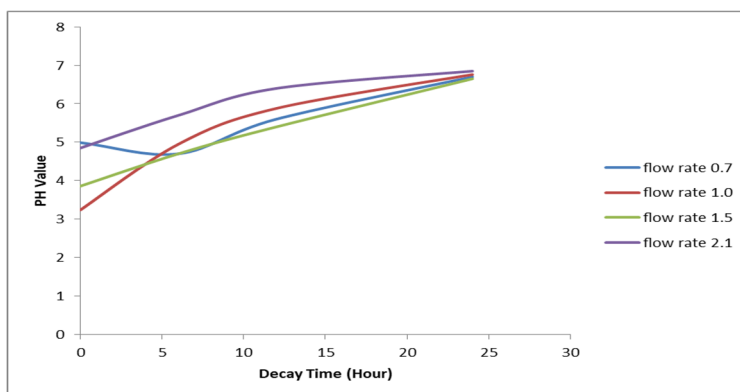


Figure 6. The relationship between the pH and the water storage time for water activated by plasma at argon gas flow rate of 0.7, 1.0, 1.5 and 2.1 l/min.

4. THE LENGTH OF THE PLASMA JET

It is good and important to know and understand the factors that control the length of a plasma jet to find the benefits of the practical use of a plasma torch. The length of the plasma jet is controlled by examining and measuring the effect of the flow rate of the argon gas. Figure 7 shows the argon gas flow rate on the length of the plasma jet. It was observed that the length of the plasma jet increased with the flow rate and that the flow rate (2.1 l/min) gave the longest length of the plasma jet.

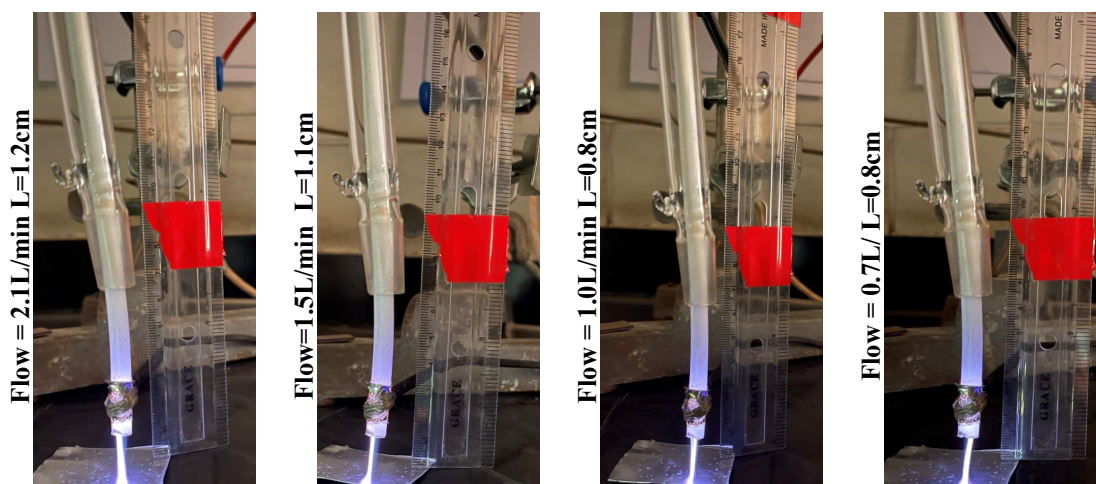


Figure 7. Effect of Ar flow rate on plasma jet length

5. CONCLUSIONS

From this research, it is possible to conclude the possibility of activating water using glow discharge plasma jet. Activating the water leads to the formation of RONS in water such as NO_2 , NO_3 , and H_2O_2 , and the concentration of these species increases with the increase in exposure time and we managed to compute actual concentrations through the use of strips measurements, whereas the majority of our predecessors relied solely on the exposure time of water to plasma. That the main factors that control the concentrations of the active species are the exposure time, as the longer the time, the greater the concentration of the active species, and the best irradiation time is 20 minutes. The second factor is the flow rate. Where the best flow rate is 0.7 l/min where this flow is laminar. The third factor is the difference in the half-lives of the NO_2 , NO_3 , H_2O_2 , where the longest-lived is H_2O_2 , followed by NO_3 , and the shortest-lived is NO_2 . The fourth factor is the possibility of converting NO_2 into NO_3 through a series of chemical reactions that are sustained by plasma. The best irradiation time is 20 min, as we get at this time the largest amount of active elements. The best flow rate is 0.7 l/min. Where the greater amount of the active species obtained 460 ppm, which is the highest quantity for different flow rates. The best storage time is 6 hours. The water temperature rises to 33°C , and the pH decreases with exposure time. When stored, the pH increases and reaches the same value as before activation after 24 hours. We deduced the half-life, the conversion of NO_3 to NO_2 and the best exposure time to water in plasma, and we found that the water is usable after a 24-hour storage period. Activating water using glow discharge plasma jet. is a simple and environmentally friendly method, so it can be used in medical and biological applications.

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ВПЛИВ ШВИДКОСТІ ГАЗУ, ЧАСУ ВПЛИВУ ТА СТАРІННЯ НА ФІЗИКО-ХІМІЧНІ ВЛАСТИВОСТІ ВОДИ, АКТИВОВАНОЇ ПЛАЗМОВИМ СТРУМЕНЕМ ТЛЮЧОГО РОЗРЯДУ

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Метою цієї роботи є вивчення того, як плазмовий струмень тліючого розряду змінює хімічні та фізичні властивості води. У цій роботі фізико-хімічні властивості води досліджувалися за допомогою плазмового струменя з газом аргон. У скляну посудину діаметром 5 см і глибиною 1 см налили 10 см³ дистильованої води. Система працювала при змінній напрузі 12 кВ і частоті 20 кГц, а час експозиції становив від 1 до 30 хвилин. Для вимірювання типів активних форм кисню та азоту (RONS), які утворюються використовувалися набори, виготовлені американською компанією Bartvation на швидкостях потоку 0,7, 1,0, 1,5 та 2,1 л/хв. Дані показали, що рівні NO₂, NO₃ і H₂O₂ були занадто високими. З часом він стає більшим і швидкість потоку зростає. Рівень pH з часом знижується, поки не досягне 3, а температура підвищується, поки не досягне 33°C. Однак pH підвищується з часом зберігання, і через 24 години вода повертається до свого природного pH 7. Кількість NO₂, NO₃ у повітрі трохи підвищується, а потім починає швидко знижуватися через 6 годин. Через 24 години концентрація близька до нуля. Звідси зрозуміло, що плазмовий струменевий пристрій тліючого розряду може створювати RONS, які можна використовувати для біологічних цілей.

Ключові слова: нетеплова плазма (НТП), постійний струм (DC), вода, активована плазмою (PAW)