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Studying the Physicochemical Properties of Water Activated by Cold Atmospheric Plasma Jet

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Abstract

This research aim is to activate water using a plasma jet that works with argon gas and to study the effect of the plasma on the physicochemical properties of the water. The 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 temperature of the produced plasma (T_e) was (1.028) eV, and its electron density (n_e) was (4.392×10^{17}) cm^{-3} at a flow rate of 1L/min. The physicochemical properties of the water are determined by determining the concentration of each of NO_2 , NO_3 , and H_2O_2 , the pH of the water, and the water temperature during exposure to the plasma. The effect of storage time on the properties of the activated water is determined after 48 hours of exposure to plasma. The RONS species were measured using kits purchased from USA (Bartovation) Company manufactured for this purpose. The pH was measured with a pH meter, and the working gas temperature was remotely measured with an IR thermometer. The concentration of the three active species increases with exposure time, but the active species NO_2 concentration decreases to zero at an exposure time of 120 minutes. The concentrations of the three components decrease with the decrease in the flow rate. The pH decreases with the exposure time until it reaches 2, and the temperature increases until it reaches 35°C. The concentration of NO_3 increases with the increase of the water storage time. In the case of H_2O_2 , the concentration increases slightly, then after 10 hours it begins to decrease exponentially, reaching a value close to zero after the passage of 48 hours. For NO_2 , its concentration begins to decrease exponentially from the start of storage and reaches a concentration close to zero after 48 hours of storage. The pH increases with the storage time, and the water reaches its natural state with a pH of 7 after 48 hours of storage. From this, it is concluded that the best storage time for PAW is ten minutes after the end of the activation process, and also that the activation time should be short, but not exceeding ten minutes.

Keywords: Plasma jet, plasma activated water, Reactive species, pH value, NO_3 , NO_2 , H_2O_2

دراسة الخواص الفيزيائية والكيميائية للمياه المنشطة بنفاث البلازما الباردة الجوية

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الخلاصة:

يهدف هذا البحث إلى تنشيط الماء باستخدام البلازما الناتجة التي تعمل بغاز الأرجون. ودراسة تأثير البلازما على الخواص الفيزيائية والكيميائية للماء. تتكون البلازما من مجهز قدرة قادر على توفير جهد متناوب عالي يصل إلى 12 كيلو فولت في شكل موجة جيبية بتردد 20 كيلو هرتز، وكانت درجة الحرارة وكثافة الإلكترونات للبلازما الناتجة $eV (1.028) T_e$ وبينما بلغت $(4.392 \times 10^{17}) n_e \text{ cm}^{-3}$ عند معدل جريان 1 L/min . يتم تحديد الخصائص الفيزيائية والكيميائية للماء عن طريق تحديد تركيز كل من NO_2 ، NO_3 ، H_2O_2 ، PH ، ودرجة حرارة الماء أثناء التعرض للبلازما، وبعد 48 ساعة من التعرض للبلازما لتحديد تأثير وقت التخزين على خصائص الماء النشط. تم قياس أنواع RONS باستخدام كتات تم شراؤها من شركة (Bartovation) USA المصنعة لهذا الغرض.

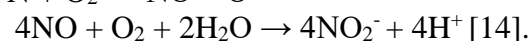
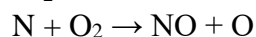
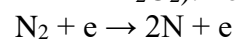
تم قياس pH بمقياس pH. وتم قياس درجة حرارة غاز العمل باستخدام ثرمومتر الأشعة تحت الحمراء عن بعد. من العلاقة بين وقت التعرض للبلازما وتركيز كل من NO_2 و NO_3 و H_2O_2 نلاحظ أن تركيز الأنواع النشطة الثلاثة يزداد بمرور الوقت ولكن NO_2 يتركب تركيزها من الصفر في وقت 120 دقيقة. ويلاحظ أيضًا أن تراكيز المكونات الثلاثة انخفضت مع انخفاض معدل التدفق. كذلك تنخفض درجة pH مع وقت التعرض حتى تصل إلى 2 و تزداد درجة الحرارة حتى تصل إلى 35 درجة مئوية. نلاحظ أن تركيز NO_3 يزداد مع زيادة وقت تخزين المياه، في حالة H_2O_2 ، يزداد التركيز قليلاً ثم بعد 10 ساعات يبدأ في الانخفاض بشكل كبير، ليصل إلى قيمة قريبة من الصفر بعد مرور 48 ساعة. يبدأ تركيزه في الانخفاض بشكل كبير من بداية التخزين ويصل إلى تركيز قريب من الصفر بعد 48 ساعة من التخزين. يزداد pH مع وقت التخزين ويصل الماء إلى حالته الطبيعية PH 7 بعد 48 ساعة من التخزين. من هذا نستنتج أن أفضل وقت تخزين لـ PAW هو عشر دقائق بعد انتهاء عملية التنشيط، وأيضاً أن وقت التنشيط يجب أن يكون قصيراً، لكنه لا يتجاوز عشر دقائق.

1. Introduction

Non-thermal plasma (NTP) is an ionized gas with a weak ionization degree and unique specifications. It has a temperature of the associated gas close to room temperature, and the average energy of the electron is about one electron volt. Non-thermal plasma can be produced by electrical discharge in gases [1]. Plasma contains electrons, ions, and neutral species, and electric field, ultraviolet and visible rays. The medical and biological applications of non-thermal plasmas using plasma-activated water (PAW) represent a modern interdisciplinary research area at the intersection of physics, chemistry, materials science, medicine, and biology [2]. 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 [3-6]. Plasma sources can be classified according to the type of feed gas, energy transferred to the plasma, shape of the voltage wave, its frequency, and the applied electric field, the temperature of the gas associated with the plasma, and the type and shape of the electrodes [7]. When using plasma in medicine, the rules of electrical safety must be adhered to, as well as the rules of protection and safety from ultraviolet radiation [8].

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), plasma jet, and coronary discharges [9-11]. The most important characteristic of these types is that the temperature of the gas associated with the plasma is close to room temperature [12]. The small size of these systems allows applications on external surfaces [13]. When the plasma touches the surface of the liquids, various chemical and physical processes take place in the interface area between the plasma and the liquid [14, 15]. These processes depend on

the type and electron energy of the plasma, and the chemical composition of the liquid [14]. When the plasma interacts with atmospheric air, which mainly contains oxygen and nitrogen before it touches the liquid surface, reactive oxygen species (ROS) such as hydroxyl OH radicals, atomic oxygen O, ozone O₃, and hydrogen peroxide H₂O₂ will be formed [16-18]. These components have a role in the process of activating the water. Also, reactive nitrogen species (RNS) such as NO₂, NO₃ and peroxy nitrite are formed [19, 20]. When these compounds reach the surface of the liquid, they 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 the formation of free radicals that contribute a lot to the applications of PAW when it is used in the medical or biological field [21]. The PAW takes place without any chemicals, so it is considered a green method. PAW has broad spectrum activity against bacteria and other pathogens. Plasma interacts with atmospheric air consisting of nitrogen and oxygen and also interacts with water vapor, and the following compounds are formed: NO₂, NO₃, H₂O₂, H₂O₂ formation comes from •OH reaction (•OH + •OH → H₂O₂). For NO₂⁻ formation includes following reactions:



This research aims to produce PAW using plasma jet that works with argon gas, to study the effect of the plasma on the physical properties of water by determining the effect of the plasma on the water temperature and the pH as a function of time, to study the effect of plasma on the chemical properties of water by determining the concentration of NO₂, NO₃ and H₂O₂ as functions of the plasma exposure time, and to check the possibility of storing the plasma activated water.

2. Experimental works

Water was irradiated by plasma jet system. Figure 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 power supply was connected to an electrode made of aluminum wrapped at the end of a glass tube with inner diameter of 5 mm placed at a distance of one centimeter from the end of the tube. The second electrode of the system, which was in the form of aluminum plate, was placed under the water container as shown in Figure 1. The other end of the glass tube was connected to an argon gas cylinder through a flow rate regulator. 10 cm 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. For the purpose of studying the effect of plasma (T_e was 1.028 eV and its electron density n_e was $4.392 \times 10^{17} \text{ cm}^{-3}$) at a flow rate of 1 l/min on water, the two variables were adopted: the exposure time, and the flow rate of argon gas (1 l/min and 0.5 l/min.). The physical and chemical properties of the water were determined by determining the concentration of each of NO₂, NO₃, H₂O₂ and also by the pH value, and the water temperature during exposure to the plasma, and after 48 hours of exposure to plasma. Where NO₂, NO₃, H₂O₂ concentrations were measured using kits purchased from USA (Bartovation) Company manufactured for this purpose. The pH was measured with a pH meter. And the temperature was measured using remotely IR thermometer.

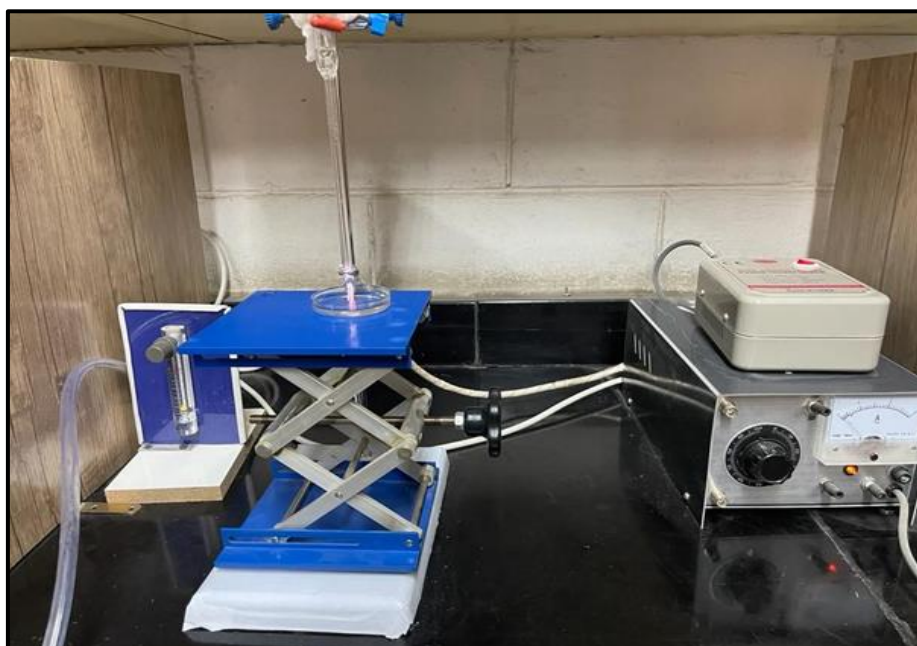


Figure1: Plasma jet system for water activation.

3. Results and discussion

Figure 2 (A and B) shows the relationship between the concentrations of NO_2 , NO_3 and H_2O_2 and the plasma exposure time at two different argon gas flow rates. From the figure, it can be noted that increasing the exposure time: the concentration of the NO_2 species reaches a maximum at exposure time of 25 minutes, after which the concentration approaches zero at 120 minutes' exposure time; the concentration of H_2O_2 increases linearly; while the NO_3 species concentration increases exponentially, which indicates that there is more than one factor that leads to the formation of the NO_3 species. It was also noted that the concentrations of the three active species decreased with the decrease in the flow rate; this is normal due to the decrease in the volume of the flowing gas.

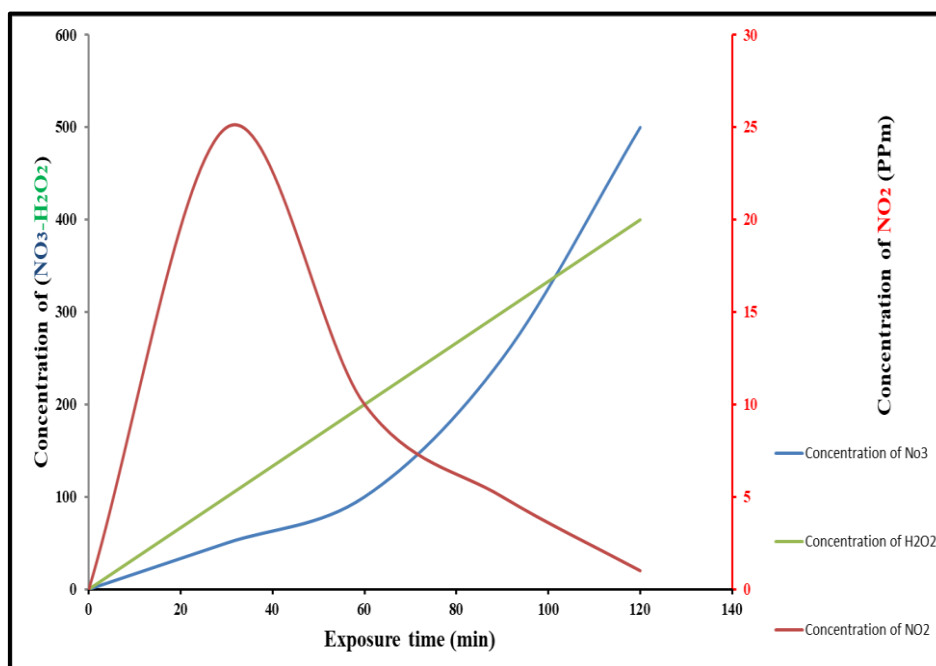


Figure 2A: The relationship between the concentration of NO_2 , NO_3 , H_2O_2 in units PPM and the exposure time at argon gas flow rate of 1 l/min

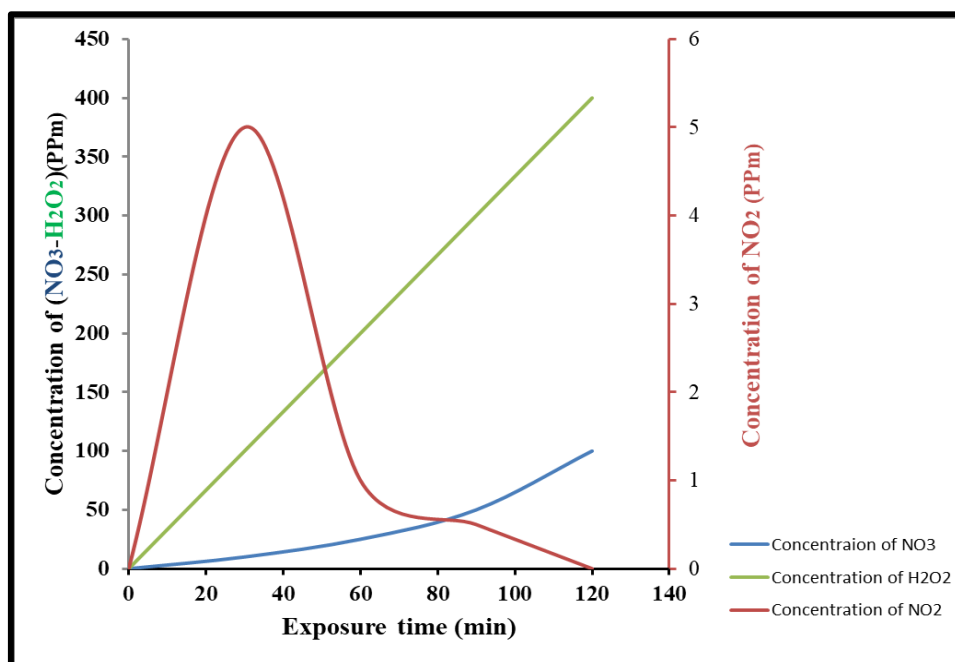


Figure 2B The relationship between the concentration of NO₂, NO₃, H₂O₂ in units PPM and the exposure time at argon gas flow rate of 0.5 l/min

Figure 3 shows the pH values as a function of exposure time at a flow rate of 1 l/min and 0.5 l/min. From the figure, a decrease in pH from 7 to 2 was noticed during 120 minutes of exposure. The same behavior was noticed for the two values of the gas flow rate. 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 the interaction of NO₃ with hydrogen, which results from the dissolution of water by the effect of plasma. Acidic solutions have a high effectiveness in reducing the effectiveness of pathogens.

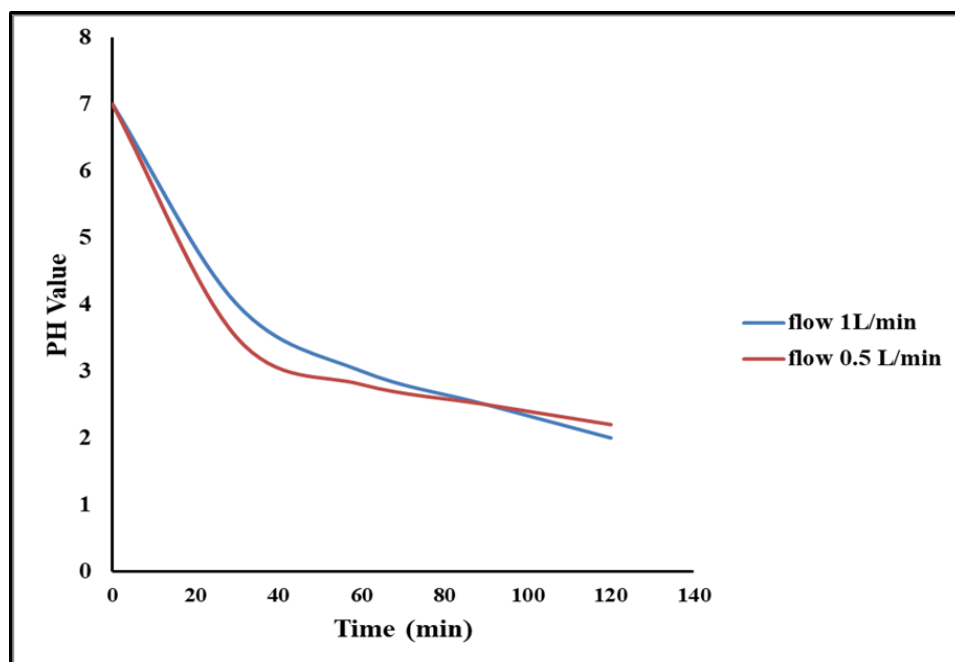


Figure 3: The relationship between the PH and the plasms exposure time at argon gas flow rate 1 and 0.5 1/min

Figure 4 shows the relationship between water temperature and plasma exposure time at a gas flow rate of 0.5 and 1 l/min. From the figure, it is clear that there is an increase in the temperature of the water, and that the highest water temperature reached was about 35 degrees Celsius, which is close to room temperature. It was also noted that the water temperature change is almost the same for the two gas flow values of flows 0.5 and 1 l/min [21].

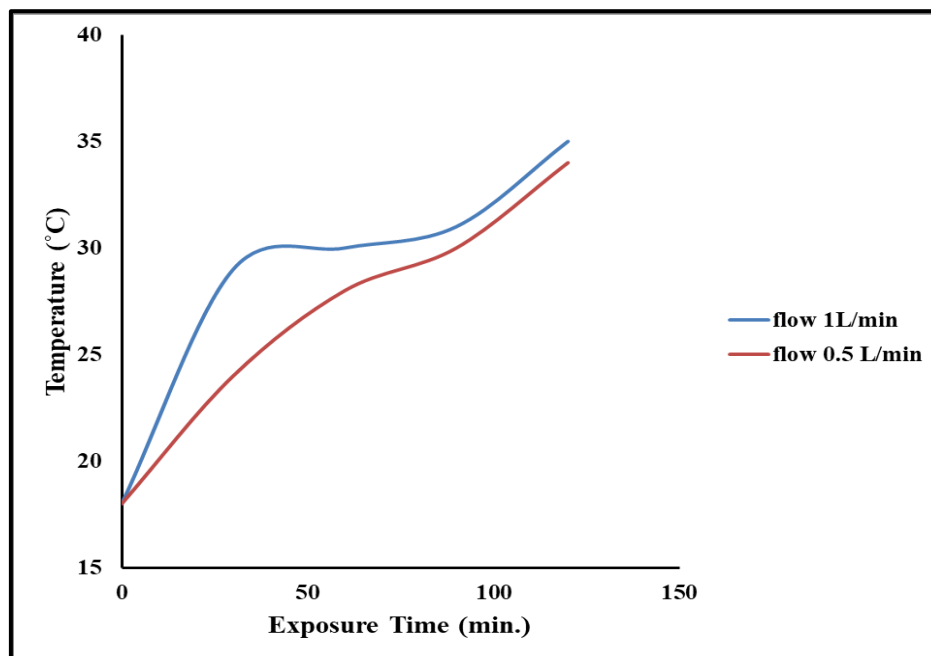


Figure 4: The relationship between the water temperature and the plasma exposure time at argon gas flow rate of 1 and 0.5 l/min

Figure 5 (A and B) shows NO_2 , NO_3 and H_2O_2 concentrations as a function of the water aging time, noting that the water was stored for 48 hours at room temperature in a tightly closed glass container. Figure 5A shows the relation at 1 l/min gas flow rate and 5B for 0.5 l/min. From Figure 5A, it is noted that the NO_3 concentration increases with the increase of the water storage time. The increase is rapid at the beginning of the storage until 24 hours, then decreases and stops at a storage time of 48 hours. In the case of H_2O_2 , the concentration increases slightly at the beginning of storage, then after 10 hours it begins to decrease exponentially, reaching a value close to zero after the passage of 48 hours. For NO_2 , its concentration begins to decrease exponentially from the start of storage and reaches a concentration close to zero after 48 hours of storage. The decrease in the concentration of NO_2 is due to its conversion to NO_3 , and the decrease in H_2O_2 concentration is due to its dissociation into H_2 and O_2 . This behavior applies in the case of 0.5 l/min gas flow rate, except that the concentration of NO_2 was close to zero at the beginning of storage, so its behavior could not be determined. From this, it was concluded that the best storage time for PAW is ten minutes after the end of the activation process, and that the activation time must be short, but not exceeding ten minutes [22].

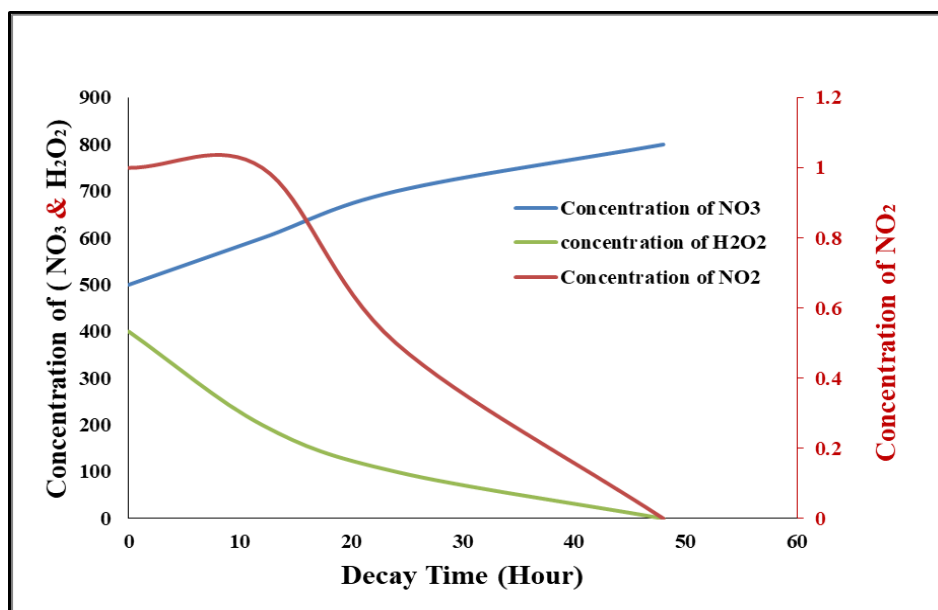


Figure 5A The relationship between the concentration of NO₂, NO₃ and H₂O₂ in units PPM and the water aging time where the water activated by plasma working with argon gas at flow rate of 1l/min.

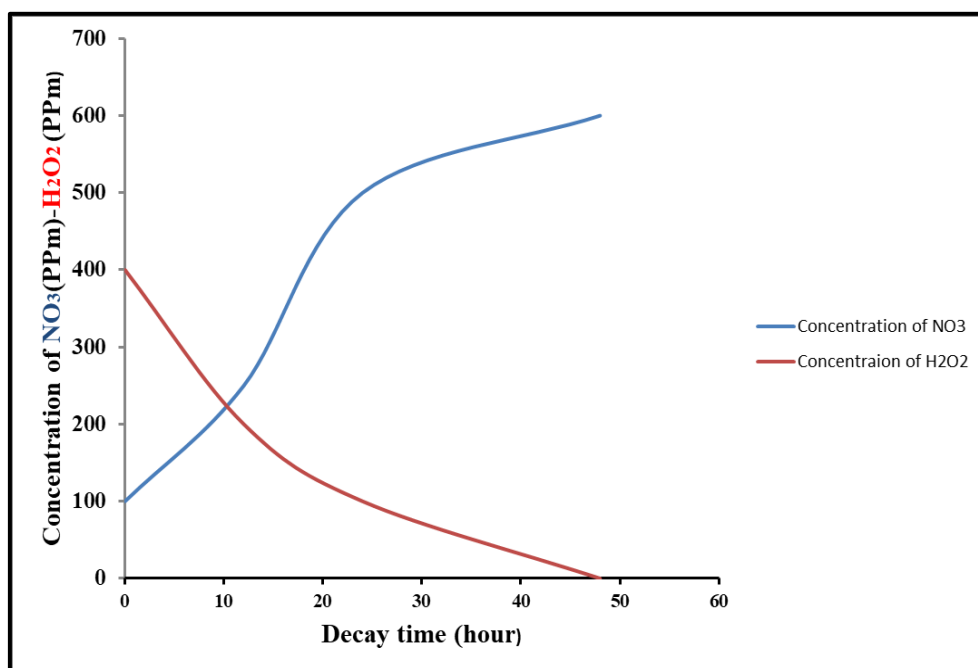


Figure 5B The relationship between the concentration of NO₂, NO₃ and H₂O₂ in units PPM and the water aging time where the water activated by plasma working with argon gas at flow rate of 0.5 l/min.

Figure 6 shows the relationship between the aging time of the PAW and the pH of the water plasma activated at a gas flow rate of 1 l/min and 0.5 l/min. It is clear from the figure that the pH increases with the storage time, and the water reaches its natural value of a 7 pH after 48 hours of storage.

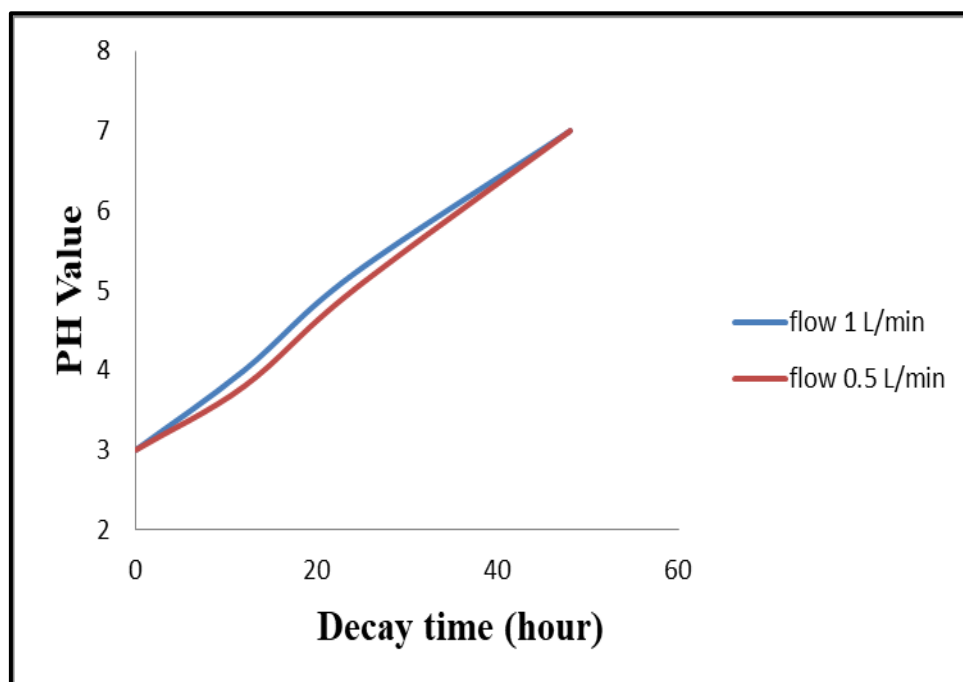


Figure 6: The relationship between the pH and the water aging time for water activated by plasma at argon gas flow rate of 1 and 0.51/min

4. Conclusions

From this research, it can be concluded that the possibility of activating water using a plasma jet and activating the water with plasma leads to the formation of RONS in water, such as NO_2 , NO_3 , and H_2O_2 . The concentration of these species increases with increasing exposure time, and the activated water can be stored at room temperature. The concentration of active species remains high after storage, and the best time for storage is 10 hours. The water temperature rises to 35 degrees Celsius, and the pH decreases with exposure time. When stored, the pH increases and reaches the value before activation after 48 hours. This makes the plasma activation process an environmentally friendly method. Plasma activated water contains active species and a low pH that gives it an acidic behavior, which makes it an effective medium for many medical and biological applications.

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