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Edited by G. D. Megersa, E. Maťaš, J. Országh, P. Papp, Š. Matejčík

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GENERATION OF REACTIVE SPECIES VIA SURFACE DIELECTRIC BARRIER DISCHARGE IN DIRECT CONTACT WITH WATER

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Surface dielectric barrier discharge (SDBD) ignited directly from the liquid electrodes at the 3-phase gas/liquid/solid interface represents a novel approach in both water and polymer surface treatment methods. This study investigates the gaseous and liquid-phase reactive oxygen and nitrogen species (RONS) generated by this discharge. The impact of the discharge power and treatment duration on the concentration of these species in both gas and liquid is explored.

1. Introduction

Plasma-activated water (PAW) in general is a type of water that has been in contact/treated with a plasma discharge. PAW has shown potential uses in numerous emerging applications, such as e.g. enhancement of seed germination, plant growth, selective treatment of cancer cells, wound healing, food preservation, inactivation of bacteria, viruses, fungi, etc. [1–4]. The versatility of PAW lies in its remarkable chemical activity, resulting from its interaction with non-equilibrium plasma. Plasma discharges in liquids or over their surface generate reactive oxygen and nitrogen species (RONS), such as radicals, ions, and excited molecules (e.g., $\cdot O$, $\cdot OH$, O_3, N_2^-, O_2^-).

Traditional surface dielectric barrier discharge (SDBD) systems generate plasma along a thin dielectric surface layer but do not directly reach the water. This limits the concentration of short-lived, highly reactive species like $\cdot OH$ and $\cdot O$ radicals. In this study, we address these limitations by employing a liquid electrode system that enables SDBD ignition directly from the liquid surface [5,6]. Although the plasma-water contact is confined to the dielectric tube's perimeter, the system is scalable and adaptable for specific applications. Besides water activation, this configuration also supports cleaning and surface treatment of dielectric materials, with potential uses in material processing and medicine. This study investigates the reactive species formation (H_2O_2, O_3, NO_2^-, NO_3^-) in tap water measured by using UV-VIS absorption spectroscopy. Additionally, Fourier transform infrared (FTIR) absorption spectroscopy was employed to measure concentrations and production yields of plasma-generated gas-phase species. The spatial evolution of O_3 in the liquid phase was also analyzed through the UV-VIS *in-situ* spectroscopy.

2. Experimental setup

To simulate the triple-phase interface (plasma-liquid-solid) under stable conditions, a thin glass test tube with a 10 mm diameter and a 0.5 mm wall thickness was used. The liquid inside the test tube served as the high-voltage electrode and was connected to a power supply generating a sinusoidal voltage waveform. The Petri dish bath, which grounded the system, completed the circuit, as depicted in figure 1. A more detailed explanation of the discharge could be found in [5] where a similar principle of the discharge was used. The high-voltage sine waveform had an amplitude range of 0 to 20 kV and could be adjusted to frequencies between 23 and 30 kHz depending on the reactor configuration and used liquids. Power was delivered to the liquid electrodes through a high-voltage resonance generator (Lifetech-300W) paired with a function generator (FY3200S-24M).

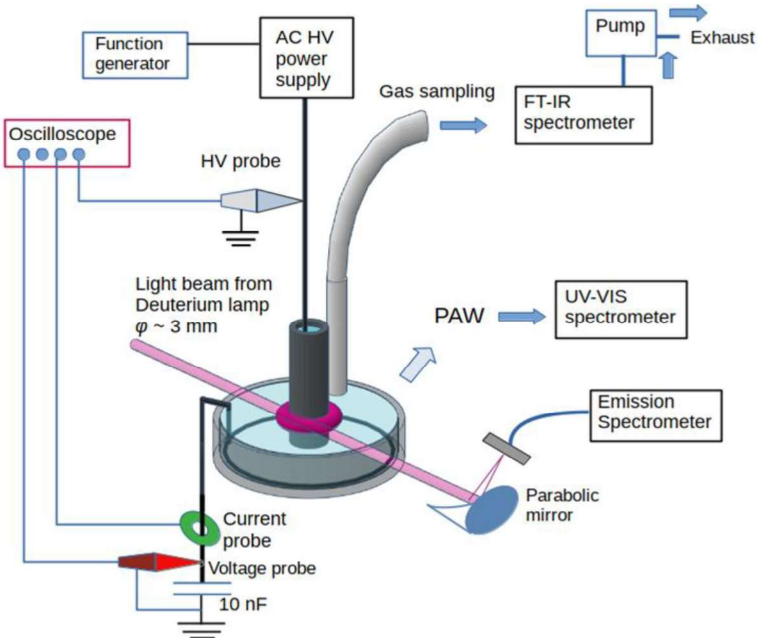

Fig. 1. Schematic of the experimental assembly for the SDBD treatment of water around the glass tube with the *in-situ* UV-VIS absorption diagnostics.

The chemical analysis of gaseous species produced by the SDBD was performed by FTIR absorption spectroscopy (Shimadzu IRSpirit-X spectrometer) using a 542 cm absorption path gas cell equipped with ZnSe windows. To determine spatial distributions of reactive species concentrations around the glass tube inserted in the treated water in the reactor, the in-situ measurements using the UV-VIS absorption spectroscopic technique were performed (figure 1). By using the UV–VIS absorption spectroscopy (Shimadzu UV-1800) the main aqueous species such as hydrogen peroxide H_2O_2, nitrite NO_2^- and nitrate NO_3^- were detected and their absolute concentrations were evaluated.

3. Results and discussion

The production of ozone (O_3) is often desirable due to its strong oxidizing properties, making it suitable for a range of applications. However, when plasma discharge interacts with a liquid, nitrogen oxides (NO_x) can dissolve, forming nitrites NO_2^- and nitrates NO_3^-, which are valuable for biomedical and agricultural purposes. The presence of water or humidity in the gas phase significantly influences the composition and concentrations of gaseous products, as well as the discharge properties and electrical characteristics. Therefore, understanding the production trends of gaseous species under various conditions is essential before employing the discharge in liquid-contact applications.

Figure 2 presents the infrared absorption spectrum of gaseous species generated by the discharge. Under the studied conditions, only O_3, N_2O, and N_2O_5 were detected. Notably, other expected species, such as NO, NO_2, HNO_2, and HNO_3, were absent from the spectra. The absence of NO and NO_2 suggests that either their concentrations were below the detection limits of the setup (approximately 7 ppm for NO and 1.5 ppm for NO_2), or they underwent rapid oxidation into N_2O_5, facilitated by O_3 [7]. The results indicate that the discharge operated primarily in an O_3-dominated mode, with negligible formation of NO, NO_2, HNO_2, and HNO_3 in the gas phase across all tested conditions.

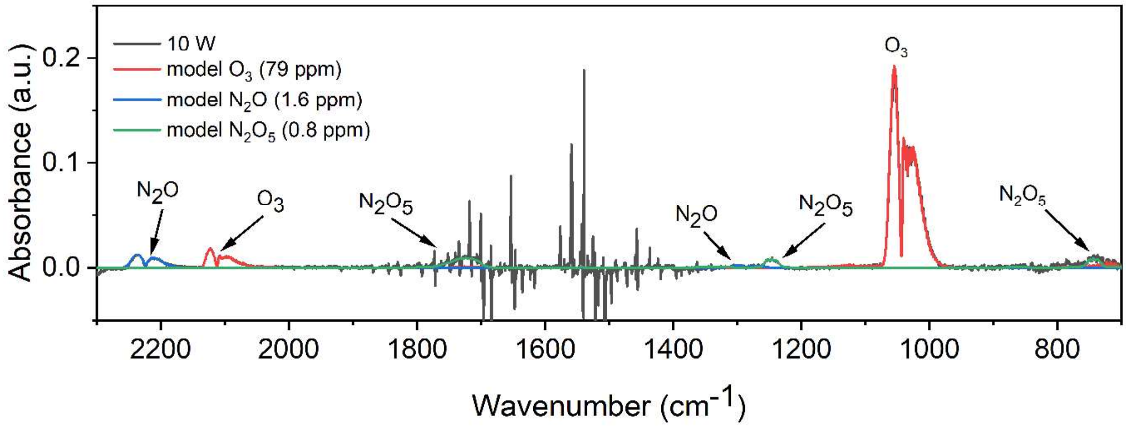

Fig. 2. Infrared absorption spectrum of gaseous products of the discharge at discharge power of 10 W. The modeled spectra of O_3, N_2O, and N_2O_5 corresponding to respective species concentrations are also presented.

Figure 3 illustrates the concentrations of RONS in tap water following the plasma treatment at varying applied powers and treatment durations. The results demonstrate that the reactor configuration highly influences RONS production. When compared to values reported in the literature, the proposed system exhibits comparable or even superior efficiency in RONS generation.

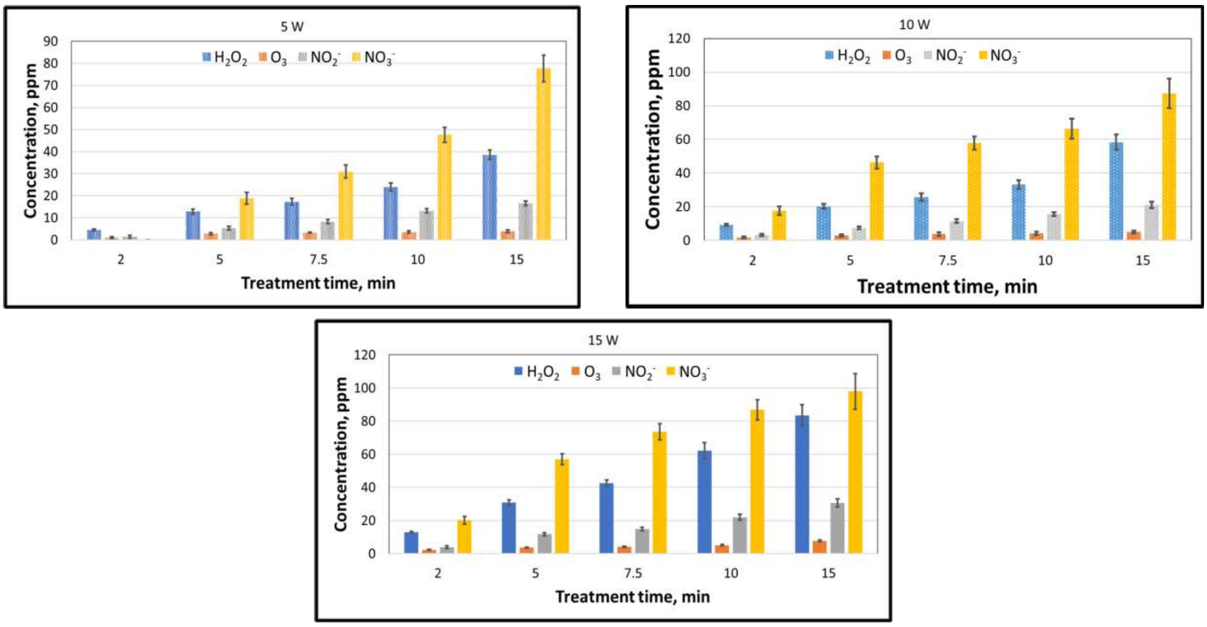

Fig. 3. Concentrations of RONS generated in the PAW outside the dielectric tube for different discharge powers.

4. Conclusions

This study employed a novel plasma setup to generate PAW, and the resulting chemical changes in both the gas and liquid phases were analyzed using various spectroscopic measurement techniques. The results showed high efficiency in the generation of reactive oxygen and nitrogen species, with ozone being the dominant product in the gas phase. The dominance of O_3 likely drives the conversion of reactive nitrogen species to N_2O_5.

The high concentration of N_2O_5 in the gas phase could explain the prevalence of nitrate ions among the RONS observed in the PAW. However, it is also possible that the observed NO3- is primarily formed from nitric acid. Since HNO_3 is readily soluble in water, it would explain its low concentrations

measured in the gas phase. Further research is needed to definitively determine the primary source of NO_3^- in the PAW.
The implementation of the surface dielectric barrier discharge (SDBD) at the gas/liquid/solid interface introduces an innovative approach to dielectric surface modification, and water treatment marking a contribution to the scientific literature with multiple environmental and biomedical applications.

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