Journal of Electrostatics 87 (2017) 284–292

Contents lists available at ScienceDirect

Journal of Electrostatics

journal homepage: www.elsevier.com/locate/elstat

Time stability of water activated by different on-liquid atmospheric pressure plasmas

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ARTICLE INFO

Article history:
Received 26 November 2016
Received in revised form
8 May 2017
Accepted 9 June 2017

Keywords:
Micro-discharge
Plasma activated water
Active species
Time stability

ABSTRACT

Two atmospheric pressure micro-jets generated in helium or argon and an air micro-arc discharge were used to obtain plasma activated water. The new water properties depend on the plasma gas, treatment time and treated volume. The higher concentration of hydrogen peroxide is associated with the argon discharge while the higher concentration of nitrogen based species and the higher modifications of pH and of electrical conductivity are associated with the air discharge. The properties of the treated water (pH and electrical conductivity) and the concentrations of hydrogen peroxide and of nitric acid show very small variations over three weeks after treatment.

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1. Introduction

Under the action of atmospheric pressure plasma (APP) water-based liquids and distilled water change their characteristics, mainly pH and electric conductivity, and acquire an important oxidative potential. The so treated liquids are named plasma activated liquids (particularly, plasma activated water – PAW). During the last decade, different types of APPs were used for activation of liquids. Dielectric barrier discharges, plasma jets, pulsed discharges, transient spark discharges, corona discharges or gliding arc discharges, generated on the liquid surface, were the most frequently used [7–11,15,18,20,23,25].

Depending on the nature of the discharge gas (argon, helium, air, oxygen or their mixture), reactive oxygen species (ROS – ozone O_3, hydrogen peroxide H_2O_2, hydroxyl radical $\cdot OH$) and reactive nitrogen species (RNS – peroxynitrite, nitrate, nitrite and the corresponding acids, nitrogen oxides NO_x) are generated in the plasma core or in the plasma-liquid contact zone being then dissolved in the liquid [3,7,8,11,14,15,18,21,23,27]. Some of them are long-lived species and the others are short-lived species [13]. They are responsible for a large variety of applications of plasmas in contact with liquids, mainly in the bio-medicine field and for reduction of environmental pollution [7–11,25,31]. The presence of nitrogen is important for the activation of water-based liquids. Generally, it enters in the discharge from ambient air as a result of diffusion and contributes to the generation in plasma of reactive oxygen and nitrogen species which are then transferred to the liquid phase [7]. Computational models developed for simulation of reactive species generation in the plasma-liquid contact region demonstrate the complexity of the physical and chemical processes taking place when a plasma is in contact with liquids [13]. The obtained theoretical results are confirmed by the experimental ones. When the plasma is generated at the surface of the treated liquid, beside the plasma gas composition and the type of applied electric field, the generation of active species process is influenced by the electrode geometry and by the gas gap (electrode-liquid distance) [3,7,13].

Water easily changes its physical and chemical characteristics under plasma exposure. However most of the studies reported a transient character of the new properties [11,14,20]. After the plasma treatment ends, the pH value starts to increase and the RONS suffer losses in terms of concentration. One study [24] assesses the importance of the storage conditions. The authors managed to control the modifications in water activity by storing it at very low temperatures (−80 °C). However at positive temperatures, the PAW suffers important modifications in time.

The current work presents a study on obtaining PAW under the action of APPs generated with helium, argon or air in the ambient atmosphere. Two different powered electrodes were used for generation of plasma in contact with the water surface: the first is a syringe needle trough which helium or argon flows as the main

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http://dx.doi.org/10.1016/j.elstat.2017.06.002
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plasma gas for generation of a small jet and the second is a metallic wire for generation of a small air glow discharge. The common characteristics of all discharges are the frequency of the applied electric field, the power transferred to the plasma and the distance between the electrode and the surface of the water. An important connection between the emission spectra of the discharges and the evolution of PAW characteristics is established and a time stability of more than 20 days is highlighted for PAW samples stored at room temperature. The paper proposes a method of generating PAW with stable properties that is very easy to store, an important aspect for future applications.

2. Experimental details

2.1. Experimental set-up

The experimental setup presented in Fig. 1 is very similar to the one presented in our previous works [1,26,30], excepting the discharge reactor. In the current experiment two different powered electrodes were used for generating the plasma in contact with water. The first one is a capillary metallic tube (syringe needle, 0.6 mm i.d.) with a free end placed at a distance of 4 mm above the treated water surface. When helium or argon as main plasma gas flows through it with a flow rate of 0.3 l min^{-1} a plasma μ-jet is generated surrounded by the ambient atmosphere. The second one is a Kanthal A-1 (Fe 71.02%, Al-5.8%, Cr-22%, Mn-0.4%, Si-0.7% C-0.08%) sharp wire (1 mm diameter and 6 mm length) fixed into a brass holder. To generate a μ-arc discharge in ambient air the wire is placed at a distance of 4 mm from water surface. The common features of all discharges are the characteristics of the applied electric field, the power transferred to the plasma (16 W) as well as the distance between the electrode and the liquid. The electrodes are powered with high sinusoidal voltage (1.7 kV, 10.2 MHz) generated with a laboratory made free-running oscillator. The second electrode of the discharge, having a floating electric potential, is represented by the treated water. The generation of a μ-jet in air is not feasible as the needle electrode overheats and melts, while the generation of μ-arcs in helium or argon is not possible in an open atmosphere. Therefore a desired comparison in which the same electrode is used with all tree gases can not be made. However the differences between the discharge types were minimized by maintaining constant the other working parameters. The inserts from Fig. 1 show the μ-jet and μ-arc discharges.

2.2. Investigation techniques and methodology

2.2.1. Plasma characterization

Optical emission spectroscopy was used for plasma diagnosis. The emission spectra of the discharges were collected with an HR4000CG-UV-NIR Ocean Optics High-Resolution Fibre Optic Spectrometer with the wavelength range of 254–965 nm. The emission atomic lines and molecular bands in the plasma emission spectra were identified with the dedicated software Spectrum Analyzer version 1.7. The software was also used for the calculation of vibrational temperature of N_2 (T_{vibN2}). The rotational temperature of the OH radical (T_{rotOH}) was estimated by finding the best fit (chi-square method) of the measured molecular spectra with the synthetic spectra generated by the LIFBASE simulation software. The excitation temperatures of electrons on atomic levels for helium and argon (T_{exc}) were estimated via the Bolzmann plot method. The groups of emission lines with the wavelengths of 801.47 - 840.82 - 842.46 - 852.14 - 912.29 nm for argon and with the wavelengths of 501.57 - 587.59 - 667.81 - 706.51 - 728.13 nm for helium were chosen for estimation of T_{exc}. The Stark broadening of the hydrogen α emission line (656.27 nm) was used to determine electron densities (n_e) based on the next formula:

$$\Delta\lambda_S = 1.78\bullet\left(\frac{n_e}{10^{17}\ cm^{-3}}\right)^{2/3}$$

The used calculation algorithm is detailed in Ref. [5]. The H_α line in the emission spectrum was fitted with a Voigt profile using OriginPro 8 software. The FWHM of the Voigt profile was corrected with the instrumental (0.5 nm), Doppler and van der Waals broadenings to obtain the Stark broadening. The van der Waals

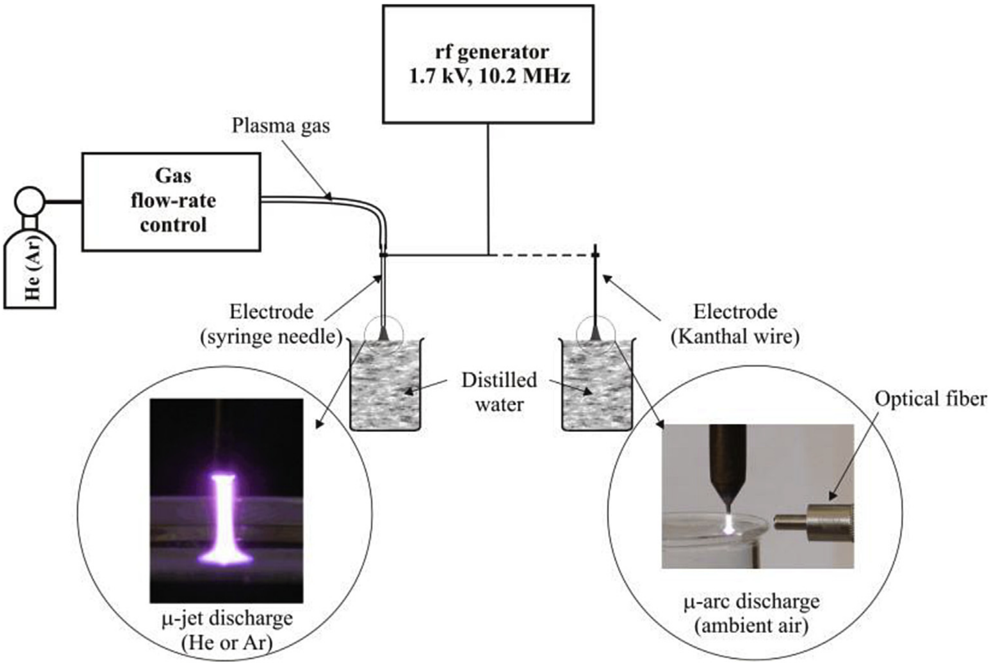

Fig. 1. Experimental set-up (the inserts show images of He μ-jet and of air μ-arc) and the positioning of the optical fiber (the discharge – optical fiber distance is 10 mm).

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