Journal of Electrostatics 91 (2018) 56–60

Contents lists available at ScienceDirect

Journal of Electrostatics

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

Inactivation of microorganisms by radical droplets from combination of water discharge and electro-spraying

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ARTICLE INFO

Keywords:
Airborne purification
Underwater discharge
Sterilization
Chemical germicide-free

ABSTRACT

We report the generation of radical droplets activated by an underwater electrical discharge and the sterilization of bacteria cells by spraying the radical droplets to open air. The plasma formed by electrical discharge in a narrow dielectric capillary tube produces atomic oxygen, atomic hydrogen, and hydroxyl radicals from the dissociation of water vapor, which is achieved through Joule heating in the narrow capillary tube. The capillary discharge electrode is placed in a water flow line, and the plasma-activated water is sprayed as fine droplets through a nozzle at 0.08 L/min. In a bactericidal test, the droplets show a sterilization effect of more than 99.99% after an exposure time of 3 h. The proposed radical droplet generator may also be useful for the elimination of airborne microorganisms.

Underwater electrical discharge [1–3] generated in a narrow capillary tube has recently attracted substantial attention as it can serve as a water purification tool for environmental cleanup activities and may be applicable to the elimination of airborne microorganisms. Furthermore, a considerable number of studies have reported results related to discharges on or in water, including water treatment or decontamination [4,5], bacteria disinfection [6,7], surgical application [8], and material synthesis application [9]. These applications are enabled by numerous active plasma chemistry reactions that are not normally observed in an aqueous solution; they ae promoted by reactive radicals, charged particles, ultraviolet light, shockwaves, and so on [1].

Discharges in and on water can initiate a variety of plasma-chemical effects, and a number of primary and secondary species can be formed by these discharges in the gas and at the gas-water interface [10]. These species can penetrate or dissolve into the water and cause chemical and biological processes in the water [11]. Among various chemical species formed by discharges in and on water, hydroxyl radical (OH), atomic oxygen, ozone, and hydrogen peroxide (H_2O_2) are the main reactive oxygen species (ROS) [12] commonly recognized to play a significant role in sterilization. Reactive nitrogen-based species (RNS), such as nitric oxide [13] and its derivatives produced with water, containing nitrites (NO), nitrates (NO_2), and peroxynitrites (NO_3), can also contribute to the sterilization process. Furthermore, the acidity of plasma-treated water increases with the production of RNS, showing synergistic effects with ROS and prolonged antibacterial activity even several days [14,15] after the exposure of water to the plasma. The transient species produced by the plasma at the gas-water interface, such as OH, NO, NO_2, and NO_3, have short lifetimes and disproportionate quickly.

The purpose of the present work is the sustainable and effective elimination of harmful airborne substances in the interior air in isolated spaces such as buildings, hospitals, and public transportation systems. We proposed a system of multiple capillary discharges connected in parallel [15,16] for the sterilization of microorganisms [1,3,17]. Herein, we report the generation of radical droplets activated by an underwater electrical discharge without any gas injection. We performed a sterilization test of *E. coli* as an example of bacteria cell by spraying the radical droplets to open air and measuring the concentration of H_2O_2 in the droplets. The proposed sterilization method may be a chemical germicide-free and toxicologically safe approach, which can be used to decrease the risk of infectious airborne diseases. Practically, our method may do away with the inconvenience to refill aqueous ROS species or H_2O_2 into a water vessel. The scaled droplet formation is beneficial for the radical or other species formation, and species lifetime or resulting disinfection.

Fig. 1 shows a schematic diagram of the apparatus for generating the underwater electrical discharge and for producing the radical mist in open air. As shown in Fig. 1, the electrical discharge in this study is formed in water without additional gas injection, the plasma-treated water exits through a spray nozzle, and the transient species of short lifetime is disproportionated quickly. Moreover, the production of RNS

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https://doi.org/10.1016/j.elstat.2017.12.007
Received 13 September 2017; Received in revised form 21 December 2017; Accepted 22 December 2017

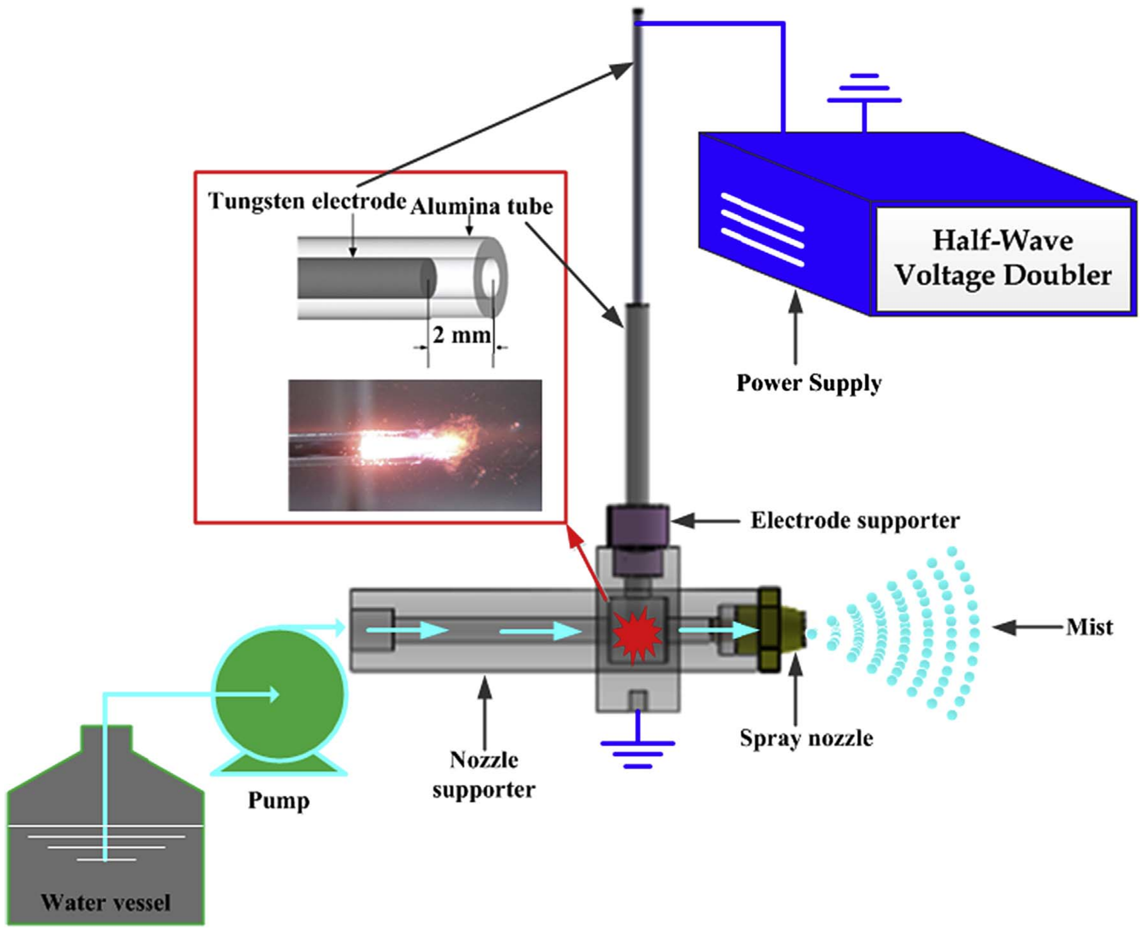

Fig. 1. Experimental set-up showing the generation of radical mist using underwater capillary discharge. The inset shows the capillary electrode arrangement and a photograph of fully discharged plasma in a vessel filled with water.

is ignorable except in the case of RNS formation from nitrogen dissolved in water. Thus, prolonged bactericidal effect in the present system can be considered to arise mainly from long-lived chemical products, which are reactive oxygen-related species.

The design and operation of the underwater capillary discharge system are briefly summarized here for completeness, although they have been described in detail in previous articles [1–3,15]. It mainly consists of a power supply, a capillary electrode, a water feeding device, and a radical mist spraying system. The power supply contains a voltage controller, an ac transformer, a capacitor, a diode, and a resistor for providing a half-wave rectification circuit, which is not shown in Fig. 1. The capillary electrode was formed by inserting a tungsten rod with a diameter of 3.2 mm into an alumina tube, as shown in the inset of Fig. 1. The assembled capillary electrode is installed in a water-flowing channel of a stainless-steel nozzle supporter with a space for discharge between the capillary electrode and the grounded nozzle supporter. Tap water with a conductivity of approximately 180 μS/cm in a vessel is introduced into the nozzle supporter through a water pump with a pumping pressure of 7 kg/cm^2. The pressurized water enters the discharge space along the flowing channel, following which capillary discharge is generated in the space, as depicted in the inset. Finally, the plasma-treated water with bactericidal chemical species is sprayed to open air *via* a metal nozzle to produce water droplets having diameters of several tens of micrometers. The photograph in the inset of Fig. 1 shows the capillary discharge plasma expanding in water visually beyond the tip of the alumina tube, emitting intense light and producing active chemical species, as well as shockwaves with distinctive acoustic sounds.

In order to visually describe the formation of radical mist in open air by the capillary discharge plasma in the water-flowing channel, selected images in Fig. 2(a) were captured after incrementally increasing the voltages between electrodes by using the voltage controller shown in Fig. 1. The distance *d* between the tips of the tungsten rod and the alumina tube was 2 mm, as shown in the inset of Fig. 1. The spray nozzle for producing fine mists in Fig. 1 was replaced with a stainless-steel cap having a 2-mm hole to visually observe the formation of radical mists, as shown in Fig. 2(a). The alumina tube covering the metal electrode is marked with a dotted line. The discharge gap between the end of tungsten electrode and the grounded cap is approximately 7 mm. Panel 1 shows the free-falling water flow from the exit of the hole at 0.08 L/min when no voltage was applied. The angle θ_{rw} between the reference line marked in white and the free-falling water column was approximately 35°. However, when the applied peak voltage $|V_p|$ was approximately 2500 V, θ_{rw} decreased to 32°, as shown in panel 2. θ_{rw} slowly decreased to almost zero as the applied voltage increased further up to $|V_p|$ = 4300, revealing a cycle mean current I_m of 0.15 A in panel 3. As shown in panel 3 of Fig. 2, the water column contains a streaming water jet with plasma and large droplets several tens of millimeters in size. Generally, the streaming jet is based on the application of a strong electric field to a fine capillary containing a liquid [18]. In this context, the Tayler cone mode in Fig. 2(b) illustrates that the accumulated charges at the water-air interface between the electrodes, in combination with a high electric field, result in a strong electric force that creates a Tayler cone, which eventually breaks into droplets. The Tayler cone mode in Fig. 2(b) represents the electro-spraying phenomenon without electrical discharge at voltages less than $|V_p|$ = 4000 V. The droplet sizes in this test significantly depend on the electric-field strength at the given water conductivity and water flow-rate. Panels