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Coupled surface dielectric barrier discharge reactor-ozone synthesis and nitric oxide conversion from air

Muhammad Arif Malik *, Karl H. Schoenbach, Richard Heller

Frank Reidy Research Center for Bioelectrics, Old Dominion University, 4211 Monarch Way, Suite 300, Norfolk, VA 23508, USA

HIGHLIGHTS

- A DBD reactor with coupled positive and negative surface streamers is reported.
- Lower power but higher chemical efficiency compared to single polarity surface-DBD.
- Multiple electrode structure of same polarity improved ozone synthesis efficiency.
- Chemical efficiency improved with gas flow through multiple chambers in series.

GRAPHICAL ABSTRACT

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ABSTRACT

A coupled surface dielectric barrier discharge is reported and compared with a surface dielectric barrier discharge with respect to the spatial distribution of the plasma streamers, the energy dissipation in the discharge plasma, the scalability of the discharges, and their efficiency for ozone synthesis and nitric oxide conversion from air. Negative streamers were found to be more effective for the chemical reactions than positive streamers. Scaling of the discharges was achieved by: (i) employing multiple interconnected electrodes in the same space and (ii) operating stacked discharge chambers in parallel in a compact configuration. The increase in efficiency caused by the two scaling methods allowed us to obtain ozone concentrations of 1–9 g/N m^3 with an energy yield of 100–70 g/kWh and nitric oxide conversions of 10–95% with an energy cost of 20–80 eV/molecule from an initial concentration of ~330 ppm in air. The results are explained on the basis of the streamer development in the two barrier discharge configurations and the results are compared with those reported in the literature.

1. Introduction

Atmospheric pressure nonthermal plasmas are being developed for a number of industrial applications [1,2]. Most of the plasma reactors for these applications are based on either the pulsed corona discharge (PCD) [3] or the dielectric barrier discharge (DBD) [4]. The PCD forms between electrode geometries where at least one electrode has a high curvature, which causes a high electric field at that electrode. Examples are pin-to-plane electrodes, wire-to-cylinder electrodes, and edge-to-plane electrodes. DBDs develop in electrode systems where the electrodes are separated by a dielectric. In the majority of cases, the electrical discharge is generated in an open gas space, i.e., generating a volume-plasma.

Placing a dielectric layer in the discharge gap between the electrodes, in the direction of the electric field, causes the electrical discharge to propagate along the solid–gas interface, i.e., generating a surface-plasma. Although there are fewer studies on surface-plasma based chemical reactors than on volume-plasmas, the results indicate that surface-plasmas are more efficient for chemical reactions [5–15]. For example, sliding discharges were found to be more energy efficient for the oxidation of volatile

* Corresponding author. Tel.: +1 757 683 2233; fax: +1 757 451 1010.
E-mail addresses: mmalik@odu.edu, MArifMalik@gmail.com (M.A. Malik).

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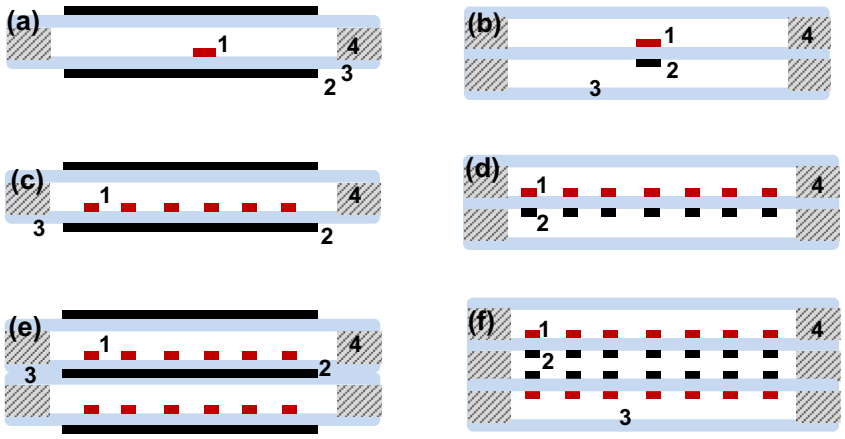

Fig. 1. Cross-section view of the reactors that are used to generate: a surface DBD with a single electrode in the chamber (a), a coupled surface DBD with a single electrode per chamber (b), a surface DBD with six parallel electrodes of same polarity in the chamber (c), a coupled surface DBD with seven parallel electrodes of the same polarity per chamber (d), a surface DBD with two stacked reactors having six parallel electrodes per chamber (e) and a coupled surface DBD with two stacked reactors having seven pairs of electrodes per reactor (f). The labels are: 1 is cathode (anode), 2 is counter electrode, i.e., anode (cathode), 3 is dielectric layer and 4 is spacer.

organic compounds (VOCs) and nitric oxide conversion from air compared to PCDs formed in the absence of the dielectric layer [8,12]. However, when sliding discharges were operated in parallel by stacking the discharge chambers, the energy density in each chamber was reduced significantly [16]. This negative effect is due to induced charges on the dielectric surface opposite to the discharge. This build-up of surface charge hampers the development of a discharge of the same polarity on that side. Extending one of the electrodes to form a conductive layer/shield on the opposite side of the dielectric successfully decoupled the neighboring discharges in the stacked chambers [16]. Further, the shield enhanced the electric field on the edges of the opposite electrode and increased the normal component of the electric field that kept the plasma firmly attached to the surface, causing a significant increase in the energy density in the shielded sliding discharge (SSD) [16,17]. A second method to overcome the effect of induced surface charges is to change the polarity of the electrodes on the opposite side of the dielectric leading to an enhancement in the energy density in such coupled sliding discharges (CSD) [18].

Placing multiple parallel electrodes of the same polarity on the same dielectric surface results in a "restricted discharge" where the propagation of the surface streamers is restricted from the electrode edges to the middle of the gap between the two consecutive electrodes. This study demonstrates, for the first time, a comparison of the electrical characteristics and efficiency of a number of chemical reactions for the "restricted discharges" in the cases of multiple parallel electrodes of the same polarity (Fig. 1c and d) and how they compare to the corresponding discharges in the cases of a single electrode per discharge chamber (Fig. 1a and b). Further, the possibility of stacking and operating multiple discharge chambers in a compact configuration (Fig. 1e and f) was also explored in this study. The efficiency of both positive streamers and negative streamers were explored. Further, the synergistic effect of discharges in two chambers placed in series was explored.

2. Experimental

Fig. 2 shows a schematic of the experimental setup. Fig. 1a–d show the cross-section views of the four reactors employed in this study (and Fig. S1 in the Supplementary file shows the corresponding top views of the electrode assemblies). The electrodes are 50 μm thick aluminum (ALF200L from Intertape Polymer Groups, USA) strips of 102 mm effective length. The effective length is defined as the portion of the electrode that overlaps the

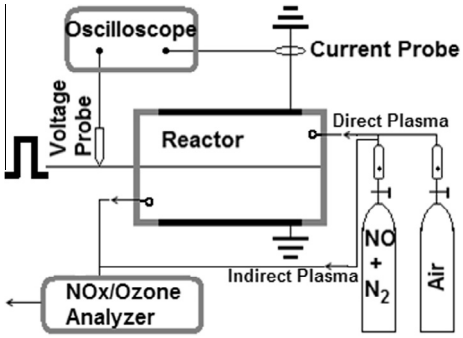

Fig. 2. Schematics of the experimental setup.

counter-electrode on the opposite side of the dielectric. The width of the first electrode was 6.4 mm and that of the second electrode was either 51 mm for surface DBDs or 6.4 mm for coupled surface DBDs of Fig. 1a and b, respectively. When multiple interconnected strips were employed as electrodes (Fig. 1c and d), the width of each strip was reduced to 1.6 mm and the gap between parallel electrodes of the same polarity was 6.4 mm. The aluminum foil was glued to a dielectric sheet made of soda glass with a thickness of 2.4 mm and lateral dimensions of 76 mm × 152 mm. A spacer frame made of silicon rubber sheet of 2.4 mm thickness, and outer dimensions of 76 mm × 125 mm was used to separate the dielectric with the electrodes from a cover plate, made of the same material and having the same dimensions as the dielectric with the electrodes. The discharge chamber, defined by the inner dimensions of the spacer frame, had a volume of 2.4 mm × 63 mm × 112 mm. The gas inlet and outlet were holes of 2.5 mm diameter on the opposite corners of the top dielectric layer connected with nylon tubes as shown in Fig. 2. Since aerodynamics caused by the discharges keeps the process gas well mixed in the chamber, neither, the direction of the gas flow, the position of the inlet/outlet holes, or the number of inlet/outlet holes had a noticeable effect on the efficiency of the chemical reactions. The side of the dielectric sheet opposite from the first electrode, in the case of the surface DBD (Fig. 1a and c), was fully covered with aluminum foil that served as the counter electrode.

A "Compact Pulsed Power Modulator MPC3000S-OP1" (Suematsu Electronics Co., Ltd., Japan) that can deliver positive high voltage (+HV) pulses of up to 28 kV peak voltage at ~150 ns duration (FWHM) and ~70 ns risetime with a repetition rate of