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Comparison between performances of surface and volume nanosecond pulsed dielectric barrier discharges for the treatment of volatile organic compounds

P. Affonso Nobrega¹, N. Blin-Simiand², B. Bournonville², F. Jorand², B. Lacour², S. Pasquiers², V. Rohani¹, F. Cauneau¹, L. Fulcheri¹

¹MINES ParisTech, PSL Research University, PERSEE - Centre for processes, renewable energy and energy systems, CS 10207 rue Claude Daunesse 06904 Sophia Antipolis Cedex, France

²Laboratoire de Physique des Gaz et des Plasmas, CNRS (UMR8578), Université Paris-Sud, Bât. 210, 91405 Orsay Cedex, France

Abstract: Non-thermal plasmas reactors are a promising alternative for treating volatile organic compounds present in air effluents. In this work, we compare the efficiencies of volume and surface nanosecond pulsed dielectric barrier discharges for the destruction of ethanol. Surface configuration was found to be much more efficient, with its characteristic energy less than half the one for volume configuration. Further, surface configuration attained more than 80 % destruction at a specific energy input of 100 J/L at 50 ppm.

Keywords: surface dielectric barrier discharges, volatile organic compounds, nanosecond pulses.

1. Introduction

Non-thermal plasmas emerge as a promising alternative for the treatment of volatile organic compounds (VOC) present at low concentrations in air effluents [1,2]. However, the energy efficiency of this technique still needs to be improved to allow a large scale adoption.

Previous experimental results suggest that two modifications of dielectric barrier discharges (DBD), used to obtain non-thermal plasmas at atmospheric pressure, may increase the energy efficiency of VOC treatment. The first is the use of nanosecond voltage pulses, with very short rise times (some hundred volts per nanosecond), instead of an AC voltage. The second is the use of a surface DBD configuration, where electrodes are placed on opposite faces of the dielectric layer and the discharge takes place along its surface.

The increased efficiency of nanosecond pulsed DBD is associated with an increased reduced electric field E/N (where E is the electric field and N the gas number density) at the discharge breakdown. A higher reduced electric field increases the production of reactive species in the discharge and, therefore, improves the VOC removal. The improved efficiency of nanosecond pulsed DBD was shown in particular for the decomposition of 2-heptanone [3] and of benzene [4].

The increased efficiency of surface DBD powered by AC-voltage was shown for the decomposition of propene [5] and the oxidation of Hg^0 [6], although the volume DBD was found to be more efficient for the decomposition of trichloroethylene by Oda [7]. Oukacine [5] suggests that the ionic wind generated by the surface discharge improves the gas mix inside the reactor, and thus helps the VOC destruction. On their side, Jiang et al. [6] remarked that ozone output was also higher in the surface DBD used to oxidize Hg^0 . That could mean that surface DBD produces

more atomic oxygen, and perhaps other reactive species, which could help VOC destruction too.

If volume and surface DBD performances have been compared under AC excitation, to the authors' knowledge no previous work has systematically compared the performances of surface and volume DBD powered by nanosecond voltage pulses. That is the goal of this work.

2. Experimental setup

Two reactors were used in this work. Both were composed of a borosilicate glass tube with an inner diameter of 10.4 mm, a thickness of 1.8 mm and a length of 110 mm. The ground electrode consisted of an aluminium foil wrapped around the glass tube. The length of the ground electrode was 50 mm. The high voltage (HV) pulse was applied on tungsten rods with 1 mm diameter and 150 mm length commercially available as electrodes for TIG welding. The difference between the two reactors were the position of these HV electrodes. In the volume DBD (VDBD) reactor, one single HV electrode was placed on the axis of the glass tube, in a typical wire-cylinder configuration. On the other side, in the surface DBD (SDBD) reactor, two HV electrodes were placed on the inner wall of the glass tube, diametrically opposed, as shown in figure 1.

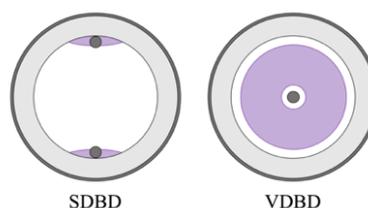


Fig. 1. Schemes of cross-sections of SDBD (left) and VDBD (right) reactors. Electrodes are shown in dark grey,

the borosilicate glass tube in light grey and the location of the plasma filaments is represented in violet.

Voltage pulses with a rise time of some 200 ns and amplitude ranging from 5.7 to 16.4 kV were applied to the HV electrodes, at repetition frequencies ranging from 100 to 32000 Hz. These pulses were generated by a home-made power source. The voltage and current signals were respectively obtained with a LeCroy PMK PVH4-2590 high-voltage probe and a T&M Research Products BNC-5-5 resistive current probe, both connected to a LeCroy Waverunner 625Zi 2.5 GHz 40 Gs/s digital oscilloscope. Typical signals are shown for the SDBD and the VDBD on figures 2 and 3, respectively. One can see that the current pulse corresponding to the discharge in the VDBD reactor is narrower and has a higher amplitude.

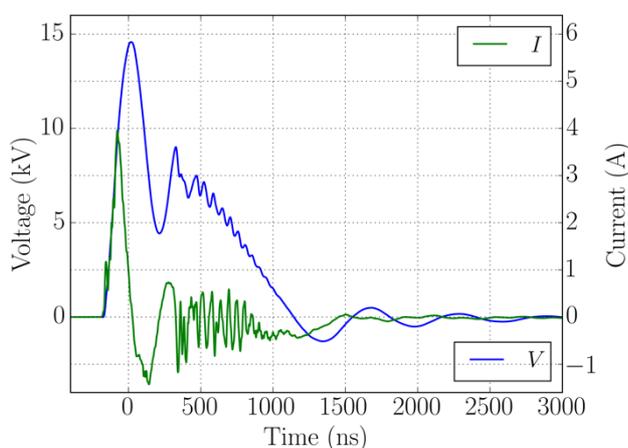


Fig. 2. Typical SDBD voltage (V) and current (I) signals. Repetition frequency: 1000 Hz.

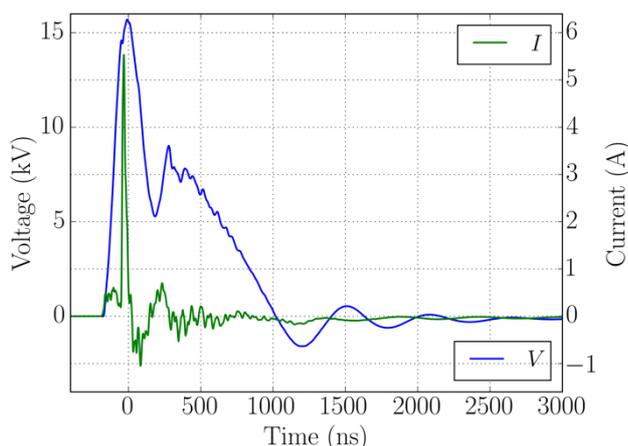


Fig. 3. Typical VDBD voltage (V) and current (I) signals. Repetition frequency: 1000 Hz.

The energy deposited in the DBD per pulse was computed from the voltage and current signals. The specific energy input (SEI) was computed by the equation:

$$SEI = E_p f / Q \quad (1)$$

where E_p is the energy deposited in the discharge per pulse, f the pulse repetition frequency and Q the inlet flow rate.

Synthetic dry air (20% O_2 , 80% N_2) was used as a carrier gas. Ethanol was used as a reference volatile organic compound. Two ethanol concentrations were used, either 50 ppm or 100 ppm. The latter was obtained from a calibrated 100 ppm ethanol-dry air mixture, while the former was obtained by mixing the calibrated mixture with pure air flow. The total flow rate was kept fixed at 1 L/min NTP. Experiments were executed at room temperature (around 25°C). The corresponding residence time in the discharge zone was 216 ms. At this flow rate, the Reynolds number is 139, corresponding to a laminar regime.

A gas chromatograph with a flame ionization detector (GC-FID Agilent 7890A) was used to quantify both ethanol and acetaldehyde outlet concentrations.

3. Results and discussion

Figure 4 shows a comparison between ethanol outlet concentration with SDBD and VDBD reactors for a 50 ppm ethanol inlet concentration. Measurements for SDBD, shown in blue, were performed with 5.7 kV, 9.5 kV and 13.3 kV pulse amplitudes. Energies per pulse were stable at 0.06 mJ, 0.50 mJ and 1.6 mJ, respectively. Measurements for VDBD, shown in green, were performed at 16.4 kV. Energies per pulse varied from 0.24 mJ to 1.81 mJ. In figure 4, we can see that the SDBD reactor is clearly more efficient in removing ethanol at those operating conditions. With the SDBD, up to 80% of the ethanol is removed at 90 J/L, against 60% with the VDBD reactor. Similar results were obtained with an ethanol inlet concentration of 100 ppm.

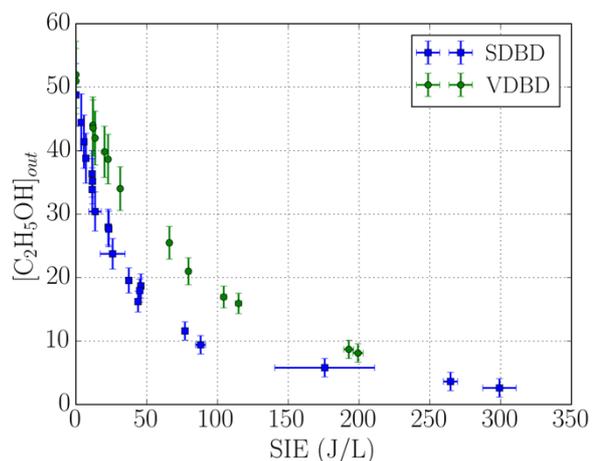


Fig. 4. Ethanol outlet concentration as a function of the specific energy input. Inlet concentration: 50 ppm. Error bars correspond to a 95% confidence interval.

In order to better quantify the difference between the performances of VDBD and SDBD reactors, we have computed the characteristic energy β for each of the two inlet concentrations analysed. To do so, we fitted each dataset with the following function:

$$[C_2H_5OH]_{out} = a \exp[-b SEI/(1 + c SEI)] \quad (4)$$

which was found to reasonably fit our data. From this fit, we obtained both the inlet concentration and the characteristic energy by the equations:

$$[C_2H_5OH]_{in} = [C_2H_5OH]_{out}|_{SIE=0} = a \quad (5)$$

$$\beta = [C_2H_5OH]_{in} \times \left(\frac{d[C_2H_5OH]_{out}}{dSIE} \Big|_{SIE=0} \right)^{-1} = 1/b \quad (6)$$

The attentive reader will note that for a low value of SIE ($c SIE \ll 1$), we obtain from equations 4 to 6 the exponential decay form:

$$[C_2H_5OH]_{out} = [C_2H_5OH]_{in} \exp(-SEI/\beta) \quad (7)$$

Table 1 provides the estimated fitting parameters for each case. The error margins provided correspond to a 95% confidence interval. Experimental data and fitted curves are shown in figure 5. Lines correspond to the fit of equation 4 to experimental data with fitting parameters provided in table 1. Error bars correspond to a 95% confidence interval.

Table 1. Estimated fitting parameters for each case.

Parameter	SDBD		VDBD	
	50 ppm	100 ppm	50 ppm	100 ppm
a (ppm)	49 ± 2	107 ± 3	51 ± 1	94 ± 3
b (10^{-2} L/J)	3.2 ± 0.4	1.8 ± 0.2	1.3 ± 0.1	0.8 ± 0.2
c (10^{-3} L/J)	9 ± 3	4 ± 2	2 ± 0.8	1 ± 2

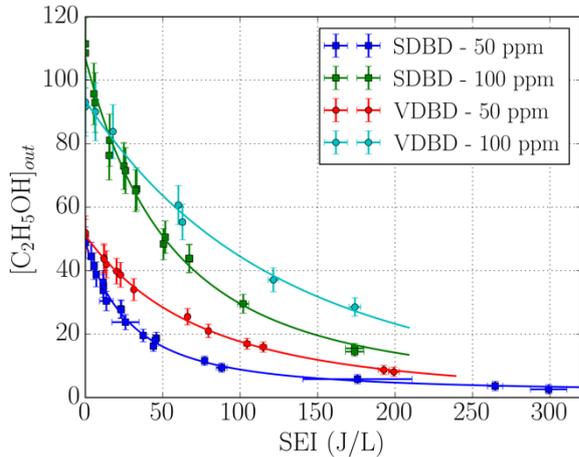


Fig. 5. Ethanol outlet concentration as a function of the specific energy input for SDBD and VDBD reactors at two inlet concentrations: 50 ppm and 100 ppm. Lines correspond to the fit of equation 4 to experimental data with fitting parameters provided in table 1. Error bars correspond to a 95% confidence interval.

Table 2 summarizes the values of $[C_2H_5OH]_{in}$ and β obtained according to equations 5 and 6, respectively. We can see that the estimated inlet concentrations are not

exactly the nominal ones. Further, the values of β confirm an increased efficiency of the SDBD reactor for both concentrations. β values for the VDBD reactor are more than twice as high as the ones for the other configuration.

Table 2. Estimated $[C_2H_5OH]_{in}$ and β for each case.

Parameter	SDBD		VDBD	
	50 ppm	100 ppm	50 ppm	100 ppm
$[C_2H_5OH]_{in}$ (ppm)	49 ± 2	107 ± 3	51 ± 1	94 ± 3
β (J/L)	31 ± 4	57 ± 8	76 ± 6	125 ± 30

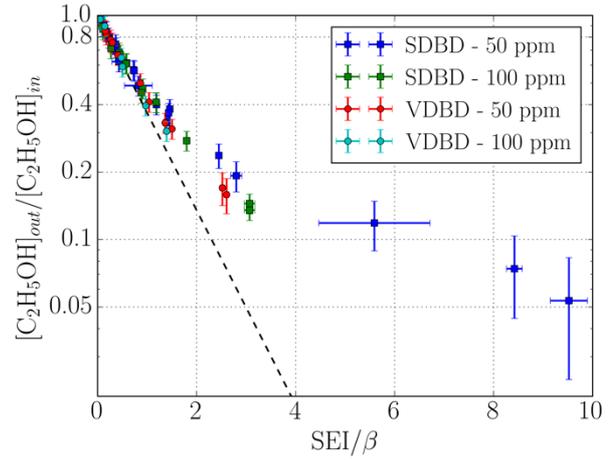


Fig. 6. Ethanol residual concentration as a function of the ratio SEI/β for SDBD and VDBD at two inlet concentrations: 50 ppm and 100 ppm. Error bars correspond to a 95% confidence interval. The dashed line corresponds to the $\exp(-SEI/\beta)$ curve.

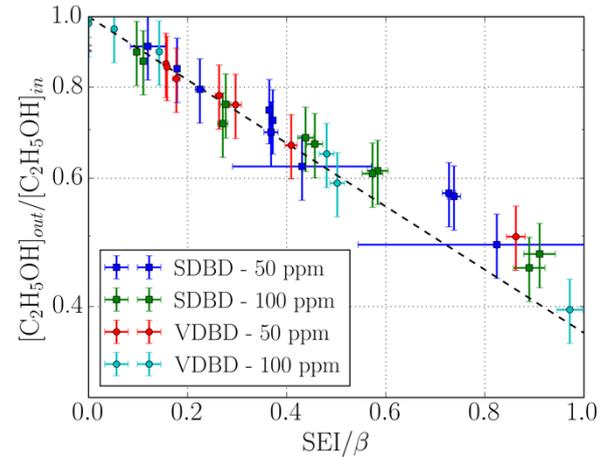


Fig. 7. Ethanol residual concentration as a function of the ratio SEI/β for SDBD and VDBD at two inlet concentrations: 50 ppm and 100 ppm and $SEI/\beta < 1.0$. Error bars correspond to a 95% confidence interval. The dashed line corresponds to the $\exp(-SEI/\beta)$ curve.

In figures 6 and 8 we use the estimated parameters given in table 2 to plot, as a function of the ratio SEI/β , the ethanol residual fraction, defined by $[C_2H_5OH]_{out}/[C_2H_5OH]_{in}$, and the acetaldehyde selectivity, defined by

$[\text{CH}_3\text{CHO}]_{\text{out}}/[\text{C}_2\text{H}_5\text{OH}]_{\text{in}}$. We can see in figure 6 that the ethanol residual concentration diverges from the exponential decay given by equation 7 for values below 60%. For values of SEI/β higher than 1, ethanol removal is much slower than the exponential decay. For the moment, we can only speculate about the reason for that. Figure 7 shows the detail of figure 6 for $\text{SEI}/\beta < 1.0$.

In figure 8 we can see that acetaldehyde outlet concentration may represent up to 15% to 20% of the ethanol inlet concentration in both SDBD and VDBD reactors. That indicates that although ethanol is destroyed, it is not fully oxidized to CO or CO₂. The presence of acetaldehyde in the outlet is in line with previous experimental results [9, 10]. In all cases, the peak in acetaldehyde selectivity is located near $\text{SEI}/\beta = 0.90$ and corresponds to an ethanol residual fraction of 50%.

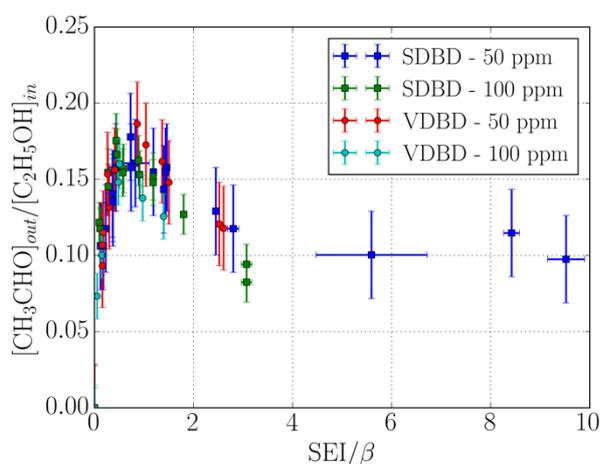


Fig. 8. Acetaldehyde selectivity as a function of the ratio SEI/β for SDBD and VDBD at two inlet concentrations: 50 ppm and 100 ppm. Error bars correspond to a 95% confidence interval.

4. Conclusions and perspectives

We have compared the efficiencies of volume and surface nanosecond pulsed dielectric barrier discharges for the conversion of ethanol in air at two inlet concentrations: 50 ppm and 100 ppm. Surface DBD was clearly more efficient at the given operating conditions, with estimated characteristic energies less than half of those estimated for volume DBD. At a 50 ppm ethanol inlet concentration, a characteristic energy of as low as 31 J/L was attained with the SDBD. Acetaldehyde selectivity was quantified, indicating that products are not fully oxidized to CO or CO₂.

In future work, the higher efficiency of the surface DBD must be confirmed for other VOCs, reactor's dimensions and operational conditions. We also aim to understand why surface DBD are more efficient. For this, additional data such as the reactors' ozone yield may be useful. Finally, we need to understand why ethanol removal is slower than the exponential decay for $\text{SEI} > \beta$, as this corresponds to a significant efficiency loss when removal increases.

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