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An SDBD plasma catalytic system for on-demand air purification.

A.J.M. Pemen, V.R. Chirumamilla, F.J.C.M. Beckers, W.F.L.M. Hoebe, T. Huiskamp

Abstract—Surface-dielectric-barrier-discharges (SDBD's) can be applied for a wide range of applications such as ozone generation, surface treatments and air-pollutants removal. An important advantage of SDBD plasma is that relatively low high-voltage pulses ($<10\text{kV}$) are needed to generate the plasma. They are effective in removing a wide range of pollutants. Despite their high energy efficiency, plasma decomposition generally results in reaction by-products and formation of ozone and nitrogen oxides. This drawback can be overcome by combining the SDBD plasma with catalysis. In this study, a novel plasma-catalytic topology is proposed, with the purpose of on-demand air purification. The main idea of the plasma catalytic reactor is that both the plasma as well as the catalytic function are configured as planar structures which are positioned in parallel to each other. The plasma is generated along a planar dielectric structure. We use a surface-dielectric-barrier-discharge (SDBD) for this purpose. Plates coated with catalytic material are positioned in parallel to the SDBD plates. The air to be treated is flushed along the plates. There are no restrictions to the type or combination of catalysts used; the catalysts and their specifications can be chosen freely. We have developed a modular plasma-catalytic SDBD reactor to handle large flows and that can be scaled-up and scaled-down easily. To energize the plasma, a SDBD power modulator was developed. The modulator is able to generate a high-voltage output pulse over an SDBD plasma load with a magnitude adjustable from 4.78 to 6.95 kV. The pulse rise time is about $1\text{ }\mu\text{s}$ and its ramp is about $6\text{ kV}/\mu\text{s}$. The energy-per-pulse then ranges from 1.1 to 17.4 mJ. The output power can be adjusted up to 48 W at a repetition rate of 5.5 kHz. The maximum possible pulse repetition rate is 22 kHz, but is limited to 5.5 kHz due to the limited current rating of the available DC power supply. The total energy efficiency of the power modulator is 68 %. A single modulator unit can power up to 2 SDBD reactor plates of $100 \times 150\text{ mm}$ size, having plasma at both sides of the plate. The operational efficiency of the developed SDBD catalytic reactor has been investigated by studying the removal of NO_x and ethylene. The removal efficiency of NO_x and ethylene are determined as a function of energy density and operational parameters such as their initial concentrations and the gas flow rate.

Index Terms—Surface-dielectric-barrier-discharge (SDBD), pulse power systems, plasma applications, plasma catalysis, air purification, NO_x removal, ethylene removal.

I. INTRODUCTION

NON-thermal plasma techniques can be effectively used for air purification [1]–[4]. They efficiently decompose volatile organic compounds, odours and other gaseous pollutants. Plasmas efficiently generate chemically active species,

Figure 1: (a-b) Conventional plasma-catalytic configurations: (a) in-plasma catalysis, (b) post catalysis. (c) New topology: planar arrangement of a SDBD plasma plate in parallel to a catalytic plate. The air to be treated is flushed along both plates.

such as radicals, excited species, ions and UV photons. Energetic plasma electrons induce chemistry by vibrational excitation, leaving the bulk gas temperature reasonably unaffected [5].

However, despite their high energy efficiency, plasma decomposition generally results in organic and inorganic (and sometimes solid) reaction by-products. Also formation of ozone and nitrogen oxides is unavoidable [6]. This by-products formation is a major drawback of plasma technology. This drawback can be overcome by combining plasma with catalysts [7]–[9]. It has been demonstrated that plasma assisted catalysis may increase the efficiency of air purification, while the formation of by-products may be significantly reduced [10]. Plasmas may promote the dissociative adsorption of reactant species at the catalytic surface and also facilitate product desorption and active site regeneration [11]–[13]. Plasma assisted catalysis will also improve the stability and life time of the catalyst, as catalyst poisoning will be reduced. Plasma is able to activate the catalyst at low (room) temperature, avoiding the need for an additional UV source (in case of a photocatalyst), and avoiding the need to heat up the catalyst as in thermal catalysis.

In existing plasma-catalytic topologies, the catalyst is positioned either inside the plasma (in-plasma catalysis, see Fig. 1(a)) or outside the plasma zone (post plasma catalysis, see Fig. 1(b)). The main disadvantage of post-plasma catalysis is that the plasma is separated from the catalyst, resulting in poor utilization of all reactive species and the UV produced by the plasma. The main disadvantages of in-plasma catalysis are that the presence of the catalyst hampers the plasma generation, and that the types of catalytic materials that can be combined with the plasma are limited.

A novel plasma-catalytic topology is proposed here (Fig. 1(c)), with the purpose of on-demand air purification. The topology proposed here is a new way of combining both the in-plasma and the post-plasma catalytic configurations. Both the SDBD catalytic reactor design, as well as the driving power

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modulator and its practical implementation will be presented. The electrical characteristics of the developed SDBD catalytic reactor will be discussed in detail. The operational efficiency of the SDBD catalytic reactor has been investigated by studying the removal of NO_x and ethylene. The removal efficiency of NO_x and ethylene are determined as a function of energy density and operational parameters such as initial concentration and gas flow rate.

II. TOPOLOGY OF THE SDBD PLASMA-CATALYTIC REACTOR

The main concept of the plasma catalytic reactor is that both the plasma as well as the catalytic function are configured as planar structures which are positioned in parallel to each other (Fig. 1(c)). The plasma is generated along a planar dielectric structure. We use a surface-dielectric-barrier-discharge (SDBD) for this purpose (see Fig. 14) [14]–[16]. Plates coated with catalytic material are positioned in parallel to the SDBD plates (Fig. ??(c)). The air to be treated is flushed along the plates. The distance between the two plates is such that the plasma is not hampered by the catalyst, but such that part of the reactive species, excited molecules and the UV generated by the plasma might be active at the surface of the catalytic plate. In this way, our new topology combines the advantages of the two conventional topologies. Since the catalytic plates are not in direct contact with the plasma, there are no restrictions to the type or combination of catalysts used; the catalysts and their specifications can be chosen freely. A SDBD reactor consists of a dielectric plate (generally ceramic), with metallic strip electrodes at both sides. Electrodes at one side are connected to (pulsed) high voltage, while the electrode strips at the other side are grounded. Plasma is generated along the dielectric surface, at both sides of a reactor plate. The catalytic plates consist of ceramic plates, coated with a catalytic material. More details of the SDBD reactor and the catalytic plates will be presented in Section IV.

The plasma plates and catalytic plates can be combined in a reactor in various ways, as shown in Fig. 2. The number of plasma plates and catalytic plates, as well as the distance between them can be varied and optimized. This also allows the reactor to be scaled up to the amount of air to be treated. A reactor can be easily adjusted for varying flow conditions, pollutant concentrations, required plasma energy density, and operating conditions. This configuration allows easy conversion of an in-plasma catalytic configuration to a post-plasma catalytic configuration just by re-arranging the plates. The order of the plates can easily be varied. Also plates with various catalytic material can be applied, allowing the use of more than one catalytic function to optimize towards various process requirements. The air flow in the reactor can be parallel to the plates (Fig. 2(a)) or perpendicular to the plates (Fig. 2(b)), though a reactor having this last configuration might have significant pressure drop. The topology is not restricted to using plate structures. Also rod type SDBD and catalytic materials, or other shapes (circular) may be used.

A catalyst provides pathways of lower activation energy for a chemical reaction; the combination of a catalyst with a

Figure 2: Various combinations of an SDBD plasma reactor (dark blue plates), and catalytic plates.

Figure 3: Basic topology of the power modulator.

plasma may yield additional synergy by promoting reactant dissociative adsorption and product desorption and regeneration of active sites. In this topology, the vibrational states can be utilized for increased surface reactions (for a short gap width between the plasma and the catalytic plates). The eventual (small amount of) heat that is produced by these states can be used to thermally activate the catalytic surface. Not only vibrational excited states, but also the ions created in the plasma can be more efficiently utilized via increased surface reactions on the catalytic surface. In case of using a photocatalytic material, the UV generated by the plasma can be used for activating the photocatalyst.

In a practical realization of this concept, it is beneficial to use a photocatalytic material such as TiO_2 as a catalyst. TiO_2 is very interesting, because of its chemical inertness, non-toxicity and stability. Band gap matching UV photon energy induces electron-deficient holes in the valence band of the TiO_2 lattice (and electrons in the conduction band), where active oxygen is produced from water and oxygen precursors. This enhances the synergetic effect between the plasma and the catalyst.

III. THE POWER MODULATOR

In order to generate the plasma, a high-voltage power modulator has been developed. We have chosen to energize the SDBD plasma by (bursts of) high-voltage pulses with microsecond duration. The applied plasma power can easily be controlled by the pulse-repetition rate. This allows on-demand air purification, for instance by sensor-based control of the pulse repetition rate. The high dv/dt of the voltage pulse slopes results in a larger discharge current, providing more intense plasma. This section gives the topology and the practical realization of the power modulator.

A. Circuit topology

The basic circuit topology of the high voltage SDBD driver is shown in Fig. 3. The circuit consists of a buffer capacitor (C) which remains continuously charged to a voltage $V_{c,0}$ by a DC power supply, a pulse transformer to step-up the voltage and a switch (Sw). The SDBD reactor has a dominant capacitive

Figure 4: Typical schematic voltage and current waveforms.

Figure 5: Circuit topology with reduced transformer model.

behaviour. The stray inductance of the transformer and the reactor capacitance (C_{sdbd}) form a resonant circuit. The reactor capacitor C_{sdbd} will be resonantly charged through the loop $C - S_1^* - S_2^* - C_{sdbd}$ after switch Sw is closed. Typical schematic voltage and current waveforms are shown in Fig. 4. The buffer capacitor C should have a value much larger than the reactor capacitance C_{sdbd} to minimize the voltage drop over C during a pulse cycle and to double the voltage over C_{sdbd} . After switch Sw is closed, the reactor capacitance is charged to a maximum voltage of $2NV_c$ (where N is the transformer winding ratio $N = \frac{N_1}{N_2}$ with N_1 and N_2 are the number of turns of the primary and the secondary winding respectively). The factor two is caused by the resonant charging under the condition that $C \gg N^2 C_{sdbd}$ [17]. Consequently, the SDBD plasma will ignite and dissipate part of the energy that is transferred to C_{sdbd} . Since the reactor is mainly capacitive, part of the energy will subsequently swing back to C . Switch Sw is subsequently opened after one period T on the zero crossing of the current. The circuit will be damped because energy will be dissipated by the discharges in the reactor. As a result of this damping, the reactor peak voltage will be (slightly) lower than the theoretical maximum value of $2NV_c$.

Figure 5 shows the circuit topology with an equivalent transformer model. All circuit values are scaled to the primary side of the transformer by applying equations Eq.(1) to Eq.(6).

$$M = k\sqrt{L_1 L_2} \quad (1)$$

$$N = \sqrt{\frac{L_2}{L_1}} = \frac{N_1}{N_2} \quad (2)$$

$$S_1^* = L_1 - \frac{M}{N} \quad (3)$$

$$S_2^* = \frac{L_2}{N^2} - \frac{M}{N} \quad (4)$$

Figure 6: Electrical circuit of the SDBD driver unit.

Figure 7: Photograph of the SDBD driver unit. The size of the board is 196 x 87 x 63 mm.

$$M^* = \frac{M}{N} \quad (5)$$

$$C_{sdbd}^* = N^2 C_{sdbd} \quad (6)$$

where M , L_1 and L_2 are respectively the mutual, the primary and the secondary inductance of the pulse transformer, k is the coupling factor, S_1^* and S_2^* are the reduced stray inductances of the primary and the secondary winding respectively, M^* is the reduced mutual inductance of the transformer, and C_{sdbd}^* is the reduced capacitance of the SDBD plate. The period time of the pulse can be estimated by Eq.(7). The capacitance C_{sdbd} is the maximum capacitance of the reactor; so the capacitance when the plasma is fully ignited. The period time (T) will in practice be smaller.

$$T \approx 2\pi\sqrt{(S_1^* + S_2^*)C_{sdbd}^*}, C \gg C_{sdbd}^* \quad (7)$$

B. Practical implementation and circuit operation

The practical implementation of the circuit is shown in Fig. 6. A photograph of the realized circuit is shown in Fig. 7. The circuit features a magnetization circuit to balance the flux swing in the transformer core and a buffer circuit to recharge capacitor C_2 (which is C in Fig. 3) via a 300 V DC power supply.

First, the transformer pre-magnetization is done in the following way. Capacitor C_m is initially charged at the beginning of a pulse cycle to approximately 60 V. First, switch Sw_1 is closed for a duration of 20 μ s to pre-magnetize the transformer core. The primary winding of the transformer is reversely

Figure 8: Typical voltage and current waveforms of the output of the pulse transformer (full cycle).

Figure 9: Magnetization current of the pulse transformer.

Figure 10: Output voltage and current waveforms of the pulse transformer.

Figure 11: Pulse energy transfer to the reactor.

biased and the magnetization current builds up linearly via (via C_m - R_m - L_m - Sw_1). Typical voltage and current waveforms of the output of the pulse transformer are given in Fig. 8. The magnetization current is shown in Fig. 9. Capacitor C_m discharges during this process. Reactor capacitance C_{sdbd} is charged to voltage $-NV_{C_m}$. Resistance R_m is included to damp the oscillation which is caused by C_{sdbd} - L_s at the start of the pre-magnetization step when Sw_1 is closed (L_s is the leakage inductance of the transformer). This oscillation can be critically damped but this will result in additional losses.

After the pre-magnetization, the pulse generation is as follows. Switch Sw_1 is opened and switch Sw_2 is closed. The reactor capacitance C_{sdbd} is subsequently charged to 7 kV in less than 1 μs (via C_2 - L_M - Sw_2 and L_s - C_{sdbd}). Note that the magnitude of the high-voltage pulse can be adjusted by means of the DC charging voltage. After charging, the freewheeling diode of switch Sw_2 conducts the reverse current to discharge the reactor. Typical output voltage and current waveforms are given in Fig. 10. The output voltage is measured with a Northstar PVM-5 high-voltage probe, while the output current is measured with a Pearson 6600 current probe. The signals are recorded on a Lecroy Wavesurfer 454 digital oscilloscope. The energy transfer to the SDBD reactor is shown in Fig. 11. The energy is determined by numerical integration of the peak power, which is derived by multiplying the voltage and current waveforms of Fig. 10. The maximum (capacitive) energy which is transferred to the reactor is 23 mJ for a 7 kV pulse. About 14.5 mJ is transferred back to C_2 or is lost in the pulse transformer. The difference (8.5 mJ) is transferred to the plasma.

Finally, the transformer de-magnetization and C_2 recharging are performed. During charging and discharging, the primary winding of the transformer is forward biased and the magnetization current reverses (see Fig. 9). The voltage over the primary winding will swing negative until C_m is able to clamp the voltage. The magnetization energy will now be transferred back

to C_m . The demagnetization current flows via the freewheeling diode of Sw_1 (see Fig. 9). The voltage level on C_m stabilizes during repetitive operation, and balances the magnetization current.

C_2 will be slightly discharged due to the discharges in the SDBD reactor. The voltage drop on C_2 will be abrupt and within $< 2 \mu s$. Capacitor C_2 will now be recharged via C_1 - L_1 - C_2 because its voltage has dropped below the voltage on C_1 . Inductor L_1 is added to limit the peak charging current through capacitor C_2 .

C. Pulse transformer design

The most critical component in the circuit is the pulse transformer. To design this component, several trade-offs must be made. The transformer should have a low stray inductance for fast pulse generation, but also a high mutual inductance to minimize the magnetization current. Low stray inductance can be obtained by minimizing the number of windings, by choosing a magnetic material with a large μ_r value and by placing the primary and secondary windings on top of each other to obtain a high coupling coefficient (k). In contradiction, a high mutual inductance can be obtained by maximizing the number of windings.

The switch Sw_2 is closed during the whole pulse period T . The magnetic flux swing $\Delta\phi$ of the transformer core can be calculated by Eq.(8), where N is the winding ratio of the transformer. The magnitude of the primary transformer voltage V_{prim} will be near to the power supply voltage V_c .

$$\Delta\phi(t) = \frac{1}{N_1} \int_0^t V_{prim}(\tau) d\tau \quad (8)$$

The flux density B in the magnetic core with cross-section A can now be calculated with $B = \Delta\phi/A$. The inductance of the primary winding can be calculated by $L_1 = \mu AN_1^2/l$, where l is the mean magnetic path length of the core and $\mu = \mu_0\mu_r$. In

Figure 12: Design of the pulse transformer.

a similar way, the inductance of the secondary winding can be calculated.

First a commercially available core material was selected, taking some of the earlier mentioned trade-offs into account. The N97 ferrite material from Epcos (type ETD59) was selected because it can operate up to a frequency of 500 kHz and has a reasonable μ_r value of 1660. The largest available E-core was selected because the high voltage winding has to fit, and a large cross-section (A) increases the primary and secondary winding inductance. The specifications of the selected core are: cross-section $A = 368 \text{ mm}^2$, mean magnetic core length $l = 139 \text{ mm}$, $\mu_r = 1660$, and saturation flux density $B_{sat} = 0.52 \text{ T}$.

A period time of $2 \mu\text{s}$ was chosen to meet the 500 kHz constraint of the magnetic material and to minimize the number of turns. A compromise was made between the required stray inductance and the number of primary turns needed to avoid core saturation. The final transformer design has a winding ratio N of 4:71. A 0.1 mm air gap was introduced in the core to decrease the residual flux which is caused by the slightly unbalanced magnetization current (Fig. 9). The air gap decreases the effective μ of the core. Also the magnetization current will increase, and primary and secondary inductance will decrease. The equivalent μ_{eq} of the core can be calculated by Eq.(9), presuming that no flux fringing occurs in the gap ($A_{core} = A_{gap}$).

$$\mu_{eq} = \frac{\mu_{core}}{1 + \frac{\mu_{core} l_{gap}}{\mu_0 l_{core}}} \quad (9)$$

A single layer high-voltage winding was constructed to minimize the risk of high-voltage breakdown. The single layer (72 turns) secondary winding is put directly on top of the coil former (Fig. 12) using 0.6 mm enamelled wire. The winding fits over the complete length of the coil former. Multiple layers of 25 μm Kapton polyimide film are wound around the secondary for insulation. A single layer of 20 mm wide copper foil is placed on top of the insulating layer at one end of the coil former. This screen has a small slit at the top and is grounded. It eliminates parasitic capacitance between the primary and the secondary winding. The 4 turn primary winding is constructed as a double layer of 15 mm wide 35 μm thick copper foil. The layers are insulated by 25 μm Kapton foil. The primary winding is placed over the screen at the end of the coil former. The

Figure 13: Output voltage and output energy-per-pulse versus the DC supply unit, measured over a SDBD plasma load with four driver units in parallel.

secondary winding is grounded at this side of the coil former. So there is a relative small voltage potential between primary and secondary at this side. The high voltage is available at the other end of the coil former. The calculated main specifications of the transformer are (measured values are given in parenthesis): $L_1 = 40.4$ (35) μH , $L_2 = 13$ (11) μH , $N = 18$ (17.8). The total stray inductance is measured by short circuiting the secondary winding, and subsequently measuring the primary inductance, and has a value of 280 nH.

D. Modulator specifications

Powered by a DC voltage range between 170 - 280 V, the modulator is able to generate a high-voltage output pulse over an SDBD plasma load with a magnitude ranging from 4.78 to 6.95 kV (Fig. 13). The pulse rise time is about $1 \mu\text{s}$ and its ramp is about 6 kV/ μs . The output voltage linearly scales with the applied DC supply voltage. The energy-per-pulse then ranges from 1.1 to 17.4 mJ. For a DC voltage of up to about 245 V, the output voltage is too low to generate plasma over the entire SDBD plate. For higher voltages, the plasma develops over the entire SDBD plate. For this reason, the plasma consumes more energy at the higher DC voltages, resulting in the change of the slope of the energy curve in Fig. 13. For the measurements presented in Fig. 13, four modulator units were connected in parallel to two parallel SDBD reactor plates. The two parallel SDBD plates have a total capacitance of 930 pF (in case of no plasma) up to 2150 pF (in case of maximum plasma power). The output power can be adjusted up to 193 W at a repetition rate of 5.5 kHz for four units (48 W for a single modulator unit). The maximum possible pulse repetition rate is 22 kHz, but is limited to 5.5 kHz due to the limited current rating of the available DC power supply. The total energy efficiency of the power modulator is 68 %, related to the energy available in buffer capacitor C . A single modulator unit can power up to 2 SDBD reactor plates of 100 x 150 mm size, having plasma at both sides of the plate.

IV. PLASMA-CATALYTIC REACTOR DESIGN

A. SDBD reactor plates

The SDBD reactor plates as used in this work, are constructed from 1 mm thick Al_2O_3 dielectric plates. The electrodes are formed by gold-coated copper tracks on both sides of the plate, so that plasma will be generated at both sides of the plates. These tracks are produced by a PCB-production

Figure 14: SDBD-plate with no plasma and with plasma generated along the dielectric surface, with a 7 kV HV pulse. Plasma is generated at both sides of the plate.

technique. The dimensions of the ceramic plate are 150 x 100 mm. The copper tracks at one side of the SDBD plate are connected to the high-voltage modulator, while the copper tracks at the other side are grounded. The copper tracks have a width of 2.3 mm and the distance between the tracks is 2.5 mm. Plasma is generated along both sides of the planar dielectric surface as in Fig. 14.

B. Preparation of catalytic plates

The same Al_2O_3 ceramic plates as described in the previous section were also used as substrate plates for the catalyst. In this work we focus on using TiO_2 as a catalyst. The catalyst plate was made by depositing TiO_2 on the ceramic plate by using the sol-gel technique where the ceramic plates were dip coated in the sol-gel [18].

A thin film of TiO_2 was deposited on the ceramic plate in the following steps.

- 1) Before the deposition process starts, the ceramic plates were cleaned with distilled water and acetone and then dried in an oven at 100°C to ensure that the surface is very clean without impurities.
- 2) Titanium (IV) tetraisopropoxide (1.77 g) is hydrolyzed with deionized water (100 ml). An ultrasonic bath was used for effective stirring during a period of 20 min at room temperature.
- 3) The resulting titanium hydroxide precipitate is separated by centrifugation. The procedure is repeated until the alcohol generated during the hydrolysis of titanium alkoxide is completely removed (at least 2 times);
- 4) The precipitate is then dissolved in 20 ml of aqueous hydrogen peroxide (15%) to get a transparent orange sol of titanium peroxo complex and subsequently diluted with water. After dilution the color of the sol changes from orange to yellow.
- 5) Poly(ethylene glycol) (PEG400) was added in order to control the morphology of the deposited film.
- 6) After mixing, the sol is aged for 5-24 h to form a gel.
- 7) The viscous Ti-peroxo complex gel is used for dip coating. For that, a cleaned Al_2O_3 substrate is dipped into the TiO_2 gel and slowly and evenly pulled out of the gel at a uniform rate of 1 mm per second. A very thin film of TiO_2 is formed on the substrate. The film is first dried in air at room temperature followed by a 2 hours drying period in an oven at 100°C . The films formed are then

Figure 15: SEM images of ceramic plate coated with a 34 nm thin layer of TiO_2 , using the sol-gel dip coating technique. (a) Is a top view and (b) is a cross-sectional view.

Figure 16: SEM images of ceramic plate coated with a 60 nm thin layer of TiO_2 , using the sol-gel dip coating technique. (a) Is a top view and (b) is a cross-sectional view.

further heated at 400°C for 1 hour in an electric furnace in air.

The thickness of the TiO_2 layer depends on the duration of the gelification of the TiO_2 sol. We have dip coated the ceramic plate after 6 hours of gelification, resulting in a TiO_2 layer with a thickness of 34 nm. Dip coating after 24 hours results in a TiO_2 layer of 60 nm thickness. The TiO_2 coated ceramic plates were characterized by scanning electron microscopy (SEM) to observe the uniformity and the thickness of the catalytic layer. The SEM images for both the 34 nm and the 60 nm TiO_2 coated plates are shown in Fig. 15 and Fig. 16 respectively. The top view SEM images in Fig. 15a and Fig. 16a are taken in the center of a plate. These two images confirm that the coating on both the 34 nm and the 60 nm TiO_2 coated plates uniformly covers the Al_2O_3 plates. Figure 15b and Fig. 16b show cross-sectional SEM images of a coated plate and these images were taken at the edge of the substrate plate.

C. The SDBD plasma-catalytic reactor

A photograph of the reactor is shown in Fig. 17. The reactor consists of a duct with rectangular cross section that holds the SDBD and the catalytic plates, and two cylindrical input and output loft structures. The length, width and height of the reactor are approximately 470 mm, 118 mm and 109 mm respectively. The inner-walls of the reactor are covered with 7 mm thick PTFE plates provided with grooves of 1.2 mm to mount the SDBD and the catalyst plates. This design can accommodate up to 20 plates in total. The reactor casing is constructed of 1 mm thick stainless steel. In order to be able to see the plasma during operation, the reactor has a glass cover at the

Figure 17: Photo of the SDBD plasma catalytic reactor.

top. High-voltage and ground connections for the SDBD plates are realized by means of metallic finger-type connectors that are fixed inside the reactor. The inlet and outlet gas connections have a diameter of 50 mm which allows flows up to 200 litres per minute. There is a provision to place additional filters (e.g. active carbon filters) both at the inlet and at the outlet side of the reactor.

V. CHEMICAL CHARACTERIZATION

To check the performance of the developed reactor, experiments were done on NO and on ethylene removal at various initial concentrations and various gas flow-rates. We have used ethylene as a model compound for VOC's. The NO and ethylene removal efficiency and the by-products formation as a function of energy density has been studied for two reactor configurations: one configuration consisting of 6 SDBD plates and one configuration with 7 catalytic plates in between and parallel to the 6 SDBD plates.

The schematic diagram of the experimental setup used to study the SDBD reactor performance is shown in Fig. 18. The flow of gas was controlled by using mass flow controllers. The gas used in the experiments discussed in this chapter consists of a mixture of synthetic air and of N_2 containing a fixed concentration of NO (1 % NO in an N_2 calibration gas). For the experiments on ethylene removal, the gas was a mixture of synthetic air and of N_2 containing a fixed concentration of C_2H_4 (1 % C_2H_4 in an N_2 calibration gas). To get the desired input concentrations of NO and of ethylene, synthetic air (with less than 3 ppm of H_2O content) is diluted with the N_2 /NO and the N_2 / C_2H_4 mixtures. The gas mixtures are dosed at room temperature and at atmospheric pressure. The treated gas from the SDBD plasma catalytic reactor was sampled at the reactor outlet by a Fourier Transform Infrared Spectrometer (FTIR, BRUKER Tensor 27) with a 20-cm optical path gas cell to measure the exit gas concentrations. Ozone concentrations were measured at the reactor outlet with a Ocean Optics HR2000 spectrometer and a Micropack D-2000 UV source. The resulting spectra from the FTIR are used to determine the varying concentrations of NO, NO_2 , O_3 , N_2O , C_2H_4 , CO and CO_2 . A pulsed-power SDBD high-voltage modulator is used to produce the discharges. A Northstar PVM-5 high-voltage probe and a Pearson 6600 current probe were used to measure the voltage over the reactor and the discharge current respectively. Their signals were recorded on a Lecroy Wavesurfer 454 oscilloscope. For all experiments, the output pulse voltage was set to 6.95 kV and the pulse repetition rate was varied from 0-300 pps to vary the plasma energy density.

Figure 18: Experimental set-up

A. NO removal in the pulsed-SDBD plasma catalytic reactor

In this section, we present results on NO removal with the SDBD plasma catalytic reactor at various initial concentrations and various flow-rates. We have studied the NO removal efficiency and the formation of by-products as a function of the plasma energy density. All the experiments in this study were performed 6 times and are reproducible within a $\pm 5\%$ error margin.

1) *Effect of NO input concentrations for a reactor configuration with only SDBD plates:* Figure 19 shows the NO and NO_x conversion as a function of the plasma energy density for various NO input concentrations and for a flow rate of 5 slm. For these results, the reactor configuration with only SDBD plates was used. It can be observed that with increasing NO initial concentrations, the NO conversion decreases. NO is completely converted for an input NO concentration of 100 ppm at an energy density of approximately 20 J/L. Whereas for 300 ppm of NO input concentration, the maximum NO conversion is 65% even at a high energy density of 120 J/L. It can be observed from Fig. 19(b) that with increasing energy density, NO_x conversion increases initially and then starts to decrease for all the input NO concentrations. This decrease in NO_x conversion at high energy densities is due to the increasing NO_2 concentrations formed from the NO. When increasing the NO initial concentrations, NO_x conversion increases up to 200 ppm and then decreases. For 300 ppm of NO input concentration, NO_x conversion happens to be negative. Due to the fact that the plasma operates in air, additional nitrogen oxides are produced from the plasma induced N_2 oxidation. According to the conservation of mass law, the observed decrease in NO_x conversion or even a negative NO_x conversion actually is due to the extra plasma source term, apparently NO_2 .

The formation of by-products for varying input concentrations of NO can be seen in Fig. 19(c). It can be observed that the N_2O concentration increases linearly with energy density. At low energy densities, the N_2O formation does not depend significantly on the input NO concentrations. But at higher energy densities, the input NO concentration has a slight effect on the N_2O formation. O_3 formation is more prominent for low NO input concentrations. With increasing NO initial concentrations, the O_3 formation decreases. For 100 ppm of

Figure 19: NO and NO_x conversion and by-products formation as a function of energy density with 6 SDBD plates at various initial concentrations and at a fixed flow rate of 5 slm.

NO input concentration, O₃ formation is observed already at low energy densities. Whereas for an input NO concentration of 300 ppm, no O₃ is observed. The ozone formation at low NO input concentration is because in this case no NO is available to consume the O atoms, as the NO is completely converted. The excess O atoms combine with O₂ to form O₃.

2) *Effect of flow-rate for a reactor configuration with only SDBD plates:* Here, the effect of flow-rate on NO and NO_x conversions and the formation of by-products is studied. The initial NO concentration was 200 ppm. The gas flow-rates were varied from 5 slm to 15 slm. The NO and NO_x conversion as a function of the plasma energy density for various flow-rates is given in Fig. 20. It is observed that the NO and NO_x conversion do not significantly depend on the applied flow rates. The gas flow of 10 slm showed minimum NO_x conversion as compared to the flow-rates of 5, 12 and 15 slm. Figure 20(c) shows that the by-product formation depends more on the energy-density than on the gas flow-rate. Both the concentrations of N₂O and of O₃ do not significantly depend on the flow-rate and increase with increasing plasma energy density.

3) *Performance of the reactor configuration with combined SDBD and catalytic plates:* To study the performance of the SDBD plasma reactor combined with catalyst, experiments were done with three reactor configurations: (i) a reactor with only SDBD plates, (ii) Al₂O₃ plates without TiO₂ coating are placed in parallel to the SDBD plates, and (iii) Al₂O₃ plates with and TiO₂ coating were placed in parallel to the SDBD plates. The experiments on these three configurations were done with an input NO concentration of 200 ppm and at a gas flow-rate of 5 slm. Figure ?? shows the NO and NO_x conversion for the three configurations. It can be observed that the NO conversion is slightly higher and that the NO_x conversion slightly lower for the configuration with only 6 SDBD plates. When the 6 SDBD plates are combined with Al₂O₃ plates with and without TiO₂ coating, the NO_x conversions are slightly improved.

It can also be noticed that the by-products formation is reduced by combining the SDBD plates with catalytic plates. When only SDBD plates are placed in the reactor, the N₂O concentration is approximately 49 ppm at an energy density of 70 J/L. When these SDBD plates are combined with catalytic plates, N₂O concentrations are reduced to 33 ppm at the same energy density of 70 J/L. A dominant reaction that is induced by the plasma is the dissociation of oxygen, and consequently ozone is one of the key by-products formed in plasma processes. In these experiments, the ozone concentration increases with increasing energy densities for all three reactor configurations. However, combining the SDBD plates with catalytic plates has a significant effect on the O₃ concentrations. When catalytic plates with the SDBD plates, O₃ concentrations were reduced from 62 ppm to 25 ppm, which is nearly 2.5 times.

Apparently, in the case that no catalyst is present in the reactor, more N radicals are available to react with NO₂ to form N₂O. This might be the reason why higher N₂O concentrations have been observed in this case. Also more O radicals are available, resulting in the higher O₃ concentrations in the case of only SDBD plasma. When catalyst plates are placed in parallel to the SDBD plates, part of the generated O radicals and of the

Figure 20: NO and NO_x conversion and by-products formation as a function of energy density with 6 SDBD plates at various flow-rates and a fixed NO input concentration of 200 ppm.

O₃ seems to dissociate into O₂ and O at the catalytic surface. So, apparently instead of performing a catalytic function, the catalytic plates are just delivering a surface where the reactions that lead to N₂O and O₃ are quenched. This might also explain why we have not observed any difference in the by-products concentrations for using Al₂O₃ plates or using TiO₂ coated plates.

B. Ethylene removal in the pulsed-SDBD plasma catalytic reactor

We also evaluated the performance of the SDBD plasma catalytic reactor for VOC removal. Ethylene was used as a model VOC compound because of its high relevance in the food industry and in floral markets. Ethylene is a relatively simple molecule compared to other aromatic VOC's and it can easily be destroyed by non-thermal plasma. Experiments were conducted at various input concentrations of ethylene, whereby ethylene removal and the CO, CO₂ selectivity is studied as a function of plasma energy density. The CO selectivity S_{CO} and the CO₂ selectivity S_{CO_2} are defined as percentage of C₂H₄ that is converted to CO and CO₂ respectively and are calculated by Eq.(10) and Eq.(11), where CO_{out} and CO_{2,out} are the reactor outlet concentrations of CO and CO₂, and where C₂H_{4,in} and C₂H_{4,out} are the reactor in- and outlet concentrations of C₂H₄. We have not evaluated the formation of by-products in detail here. Input concentrations were varied from 50 ppm to 250 ppm at an interval of 50 ppm. The gas flow-rate was 5 slm. Experiments were done for three reactor configurations: (i) a reactor with 6 SDBD plates, (ii) a reactor with 6 SDBD plates and 6 uncoated Al₂O₃ plates, and (iii) 6 SDBD plates and 6 Al₂O₃ plates coated with TiO₂.

$$S_{CO} = 100 \frac{CO_{out}}{2(C_2H_{4,in} - C_2H_{4,out})} \quad (10)$$

$$S_{CO_2} = 100 \frac{CO_{2,out}}{2(C_2H_{4,in} - C_2H_{4,out})} \quad (11)$$

For the reactor with only SDBD plates, it can be observed from Fig. 22(a) that for all the input concentrations 100% conversion can be realized with increasing energy density. This complete removal of ethylene was also reported in [19] and [20]. With increasing input concentrations, the conversion of C₂H₄ decreases. 90 % conversion of C₂H₄ was achieved at an energy density 10.5 J/L for an input concentration of 50 ppm, whereas 76 J/L is needed for 90 % removal at 250 ppm. The selectivity towards CO and CO₂ as a function of energy density is shown in Fig. 22(b). With increasing energy density, the selectivity towards CO and CO₂ is increased. The selectivity towards CO is higher than towards CO₂. This means that the C₂H₄ is converted more to CO and further oxidation of CO to CO₂ is limited. Aerts *et al.*, [20] suggested a detailed destruction pathway for C₂H₄ with dry air and humid air in their work on gas purification by non-thermal plasma. The destruction pathway for ethylene in dry air can be summarized by the following four main steps:

- 1) C₂H₄ collides with N₂ metastables and with O atoms to produce vinyl radicals (C₂H₃) and acetylene molecules

Figure 21: NO and NO_x conversion and by-products formation as a function of energy density with catalytic plates placed alternatively to the SDBD plates in a parallel arrangement at an input NO concentration of 200 ppm and gas flow-rate of 5slm.

(C₂H₂) through the following reactions:

- 2) C₂H₃ is oxidized by O₂ to form CHO radicals and HCHO. Formaldehyde is observed as by-product at low energy densities.

- 3) C₂H₃ may also reacts with O to form CO and CH₃ or C₂H₂ and OH. The C₂H₂ exhibits a high bond energy and also high degree of reactivity. Although its reactions with electrophilic species like OH are strongly exothermic, the formation of the intermediate pi-complex is a slow, rate determining process. Therefore, C₂H₂ yet can be observed as product.

- 4) CHO is further oxidized by O, O₂ and OH to form CO and CO₂

Thus, CO, CO₂, HCHO and C₂H₂ are the main by-products that are formed from ethylene destruction. Of these by-products, with the diagnostics we have used in this study, we were only able to quantify the CO and CO₂ formation. With increasing input concentration, the selectivity towards CO and CO₂ is observed to decrease as can be seen in Fig. 22(b). This decrease in selectivity is due to the lower conversion of C₂H₄ itself at higher input concentrations. The maximum CO selectivity that is observed here is 60% for an input concentration of 100 ppm. The selectivity towards CO₂ is higher for low concentrations because at low concentrations, C₂H₄ is completely converted to CO at low energy densities and thus the remaining energy is used to oxidize CO to CO₂. Thus CO is observed as a main by-product for all the concentrations.

Due to its toxicity, CO and formaldehyde are not a preferred by-product. Thus we need higher energy densities or a longer residence time as mentioned by [19] to oxidize CO to CO₂ and to convert the formaldehyde to formic acid and CO_x. Combining plasma with a catalyst is another option to increase the selectivity towards CO₂. We tried to evaluate the third option by combining SDBD plates with TiO₂ coated catalytic plates and studied the conversion of C₂H₄, the selectivity towards CO and CO₂ and other by-products such as O₃ and NO_x. The effect of this on the C₂H₄ conversion and on the CO and CO₂ selectivity can be seen in Fig. 23. It is noticed that the C₂H₄ conversion is slightly higher when only SDBD plates are used, as for the situation that the SDBD plates are combined with the catalytic plates. On the other hand, the selectivity towards CO₂ is noted to be slightly higher for the combined SDBD and catalytic plates. Though the difference between the two configurations

(a) C_2H_4 conversion (b) CO and CO_2 selectivity

Figure 22: C_2H_4 conversion and CO, CO_2 selectivity as a function of energy density with 6 SDBD plates at various C_2H_4 input concentrations with the μs pulsed SDBD driver.

is quite small, it is notable that the catalytic plates help in improving the selectivity of the process. With the TiO_2 sol-gel method that we have used to coat the Al_2O_3 plates, we were only able to obtain a TiO_2 layer thickness of 60 nm. It will be interesting to study the performance of the reactor with a thicker TiO_2 layer which is expected to have higher catalytic activity. Since all experiments were done in synthetic air, also NO_2 and O_3 could be observed as by-products, as shown in Fig. 23(c) and Fig. 23(d) respectively. We have not observed N_2O concentrations for all the three reactor configurations and at all the applied plasma energy densities. The NO_2 concentration increases with increasing energy density. Harling *et al.*, [21] describe a novel method for enhancing the destruction of environmental pollutants by the combination of multiple plasma discharges. They also report that they have not observed any NO and NO_2 concentrations which is in contradiction to our results. Aerts *et al.*, [20] also reported very low NO_2 concentrations. This may be because of the high energy densities that they have used, so that the NO_2 is likely further converted to N_2O and HNO_2 . The SDBD plates combined with catalytic plates showed lower NO_2 concentrations and O_3 concentrations.

(a) C_2H_4 conversion (b) CO and CO_2 selectivity (c) NO_2 concentration (d) O_3 concentration

Figure 23: C_2H_4 conversion and CO, CO_2 selectivity and NO_2 and O_3 concentrations as a function of plasma energy density with and without catalytic plates placed in parallel to the SDBD plates. Input C_2H_4 concentration is 200 ppm and gas flow rate is 5 slm.

VI. CONCLUSIONS

Surface-dielectric-barrier-discharges (SDBDs) have gained importance recently due to their ease of generation and their wide range of applications, such as ozone generation, surface treatments and air-pollutants removal. In this paper, we introduce a modular plasma-catalytic surface-dielectric-barrier-discharge (SDBD) reactor to handle large flows, and which can be scaled-up and scaled-down easily.

- To energize the SDBD plasma, a power modulator was developed to generate high-voltage pulses upto 7 kV with microsecond duration.
- Such a power modulator can energize two SDBD reactor plates. We have connected 3 of such power modulators in parallel to power up to 6 SDBD plates.
- The applied power can be controlled by the pulse-repetition rate.
- The electrical and the reactor characteristics of the developed SDBD reactor have been determined by experiments.

The reactor performance was analyzed by conducting experiments on the removal of NO_x and of ethylene at various input concentrations and various flow-rates and by studying the conversion rate and the formation of by-products.

- It was found that with increasing NO initial concentrations, the NO and the NO_x conversion decreases.
- We have observed negative NO_x conversion for an input concentration of 300 ppm NO.
- Also with increasing gas flow-rate, the NO and NO_x conversion decreases. Thus higher conversion is achieved at longer residence time.
- To evaluate the VOC removal performance, experiments were done at the removal of ethylene. 100% conversion of C_2H_4 was obtained for all the input concentrations.
- With increasing input concentrations, the selectivity towards CO and CO_2 is observed to decrease. This decrease in selectivity is due to the lower conversion of the C_2H_4 itself.

To reduce the by-products formation, we combined the SDBD plasma reactors with a catalyst.

- To do so, Al_2O_3 plates without a TiO_2 coating and with a TiO_2 coating were placed alternatively to the SDBD plates in a parallel arrangement.
- This resulted in a marginal improvement of the NO_x conversion, though the formation of by-products is reduced. Observed O_3 outlet concentrations in the NO_x removal experiments were reduced from 62 ppm to 25 ppm, which is nearly 2.5 times, by combining the SDBD plasma with the catalytic plates.
- Combining the SDBD plasma with a catalyst in the way we describe in this paper is an interesting option to increase the selectivity of VOC removal towards CO_2 .
- This selectivity is notably higher when the SDBD plasma is combined with catalytic plates, thus slightly improving the selectivity of the VOC removal process.
- With the sol-gel method that we have used here to coat the Al_2O_3 plate with TiO_2 , we have managed to obtain a TiO_2 layer with a thickness of 60 nm.

- It will be interesting to study the performance of the reactor with an increased layer thickness of TiO_2 which is expected to have increased catalytic activity.

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