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Application of PlasTEP standard 1: Treatment of propene and nitrogen oxides with single stack DBD reactor

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1. Experimental conditions

Experimental setup

The plasma is generated by DBD (dielectric barrier discharge) stack reactor. The stack reactor consists of dielectric plates and metal mesh electrodes which are alternate stacked. The electrode are alternating connected to high voltage and ground potential. The concept is used since it enables gas treatment with low back pressure¹. Furthermore, due to its simple design it is easy scalable with gas flows. For laboratory studies with small gas flows a special stack reactor is used. It consists only of one barrier layer and two metal mesh electrodes (high voltage and ground electrode).

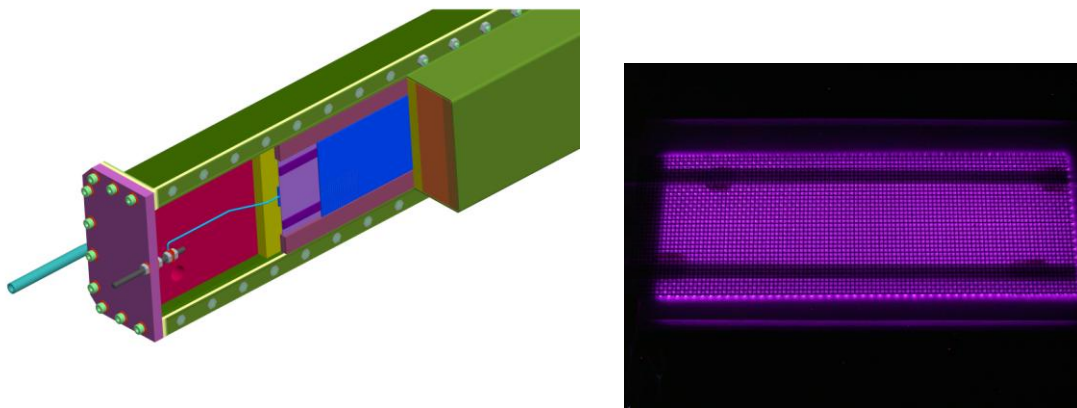


Figure 1: DBD single-stack reactor for treatment of exhaust gases; plasma in operation in open air

The flow cross section is $7.4 \cdot 10^{-5} \text{ m}^2$ and the length of the active plasma zone is 0.12 m. The plasma reactor is situated in an electric tubular furnace. The gas to be treated is supplied by gas cylinders and fed by a heated

¹ S. Müller, R.-J. Zahn; Contributions to Plasma Physics 47 (2007) 520-529



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pipe system containing mass flow controllers and a humidifier unit. The gas temperature is measured before, inside and after the oven. The concentrations of the species are monitored by an FTIR-spectrometer with a spectral resolution of 0.5 cm^{-1} . The temperature of the gas cell is set to 185°C . The reactor is energised by sinusoidal high voltage with a frequency of 400 Hz. The power dissipated into the plasma is measured by a digital oscilloscope via current-voltage-oscillography.

Gas mixture

The composition of the gas mixture which was provided by a gas mixing system using mass flow controllers is given in Table 1. The gas composition is strictly oriented on the PlasTEP-standard. The values calculated and set agree very well with the realised values except the water content which is due to condensation effects lower than the value set. Non-agreement of NO_2 is due to available test gas mixture.

component	PlasTEP-standard	value set	value measured
NO	400 ppm	400 ppm	396 ppm
NO_2	21 ppm	55 ppm	45 ppm
CO	100 ppm	100 ppm	93 ppm
CO_2	5 %	5%	5.3%
C_3H_6	500 ppm	500 ppm	435 ppm
O_2	10 %	10%	-
H_2O	5 %	5%	1.8%
N_2	balance	balance	-

Table 1: Concentrations of the gas components

The total gas flow is 600 L/h at room temperature and the gas temperature in the plasma reactor is 65°C and kept constant during the measurement. The gas velocity in the plasma reactor is 2.25 m/s which corresponds to a retention time of 53 ms or a gas hourly space velocity (GHSV), which is defined as the gas flow divided by the volume of the reactor, of about 19 s^{-1} .

2. Results and discussion

The concentrations of the species monitored are given with respect to the specific energy density (SED) which is defined as the power dissipated into the plasma divided by the gas flow and acts as the similarity parameter. Plasma power was measured via current-voltage oscillography. The method was checked with Lissajous-figure measurements.

Propene and carbon monoxide

The volatile organic compound (VOC) treated in this experiment is propene (C_3H_6). The concentration of propene with respect to the SED is given in Figure 2. At low SED-values the propene concentration is unaffected by the plasma activity. High values of $\text{SED} > 100 \text{ J/L}$ lead to a reduction of the propene-content of about 200 ppm. This corresponds to a production of carbon monoxide (CO) of about 600 ppm. Taking the



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number of carbon atoms into account it could be concluded that the propene is converted into carbon monoxide² but other experiments showed that CO is also produced in gas mixtures not containing propene. By now CO₂ is identified to be the CO-source but this must be verified further in additional studies.

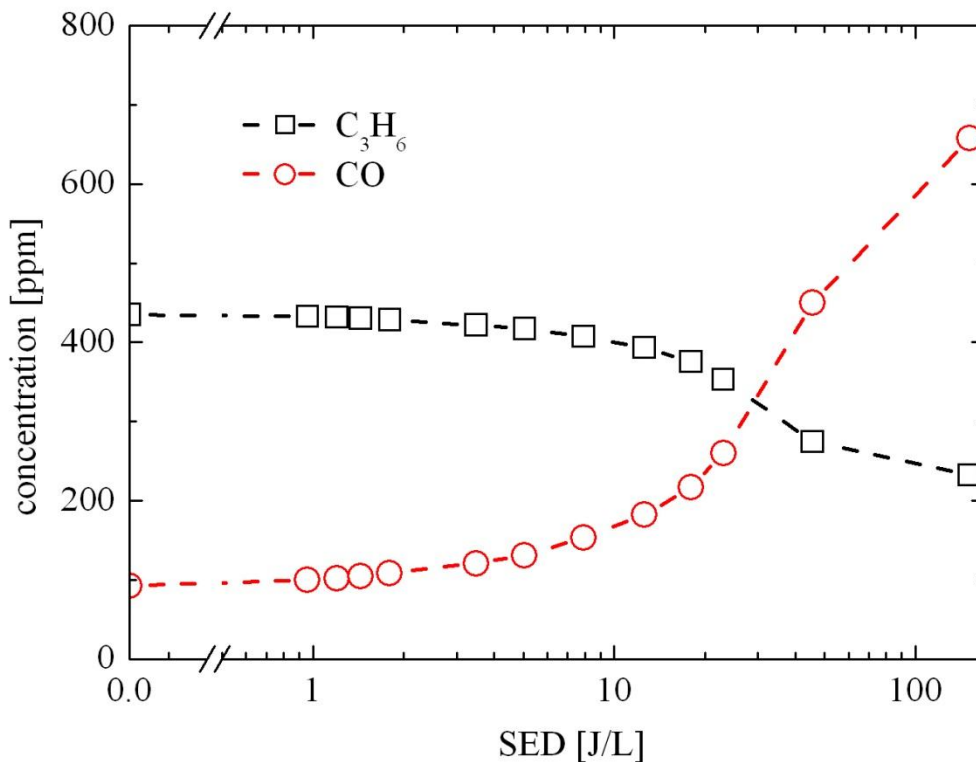


Figure 2: Concentrations of carbon monoxide and propene with respect to the specific energy density

Nitrogen oxides

In Figure 3 the concentrations of the nitrogen oxides are given. With the SED increasing the NO-content decreases and the NO₂-content increases. The NO_x-content, which is defined here as the sum of NO and NO₂, remains almost constant. This shows that NO is mainly converted into NO₂ due to oxidation by the plasma³. The comparison of the FTIR-spectra of the untreated and the treated gas mixture (SED = 150 J/L) and the search for undetected species gives that there are no species which are not monitored. This shows that the small reduction of the NO_x-content at the high SED-values is due to conversion of NO_x to components not detectable by FTIR-spectroscopy (N₂ and O₂).

² Lin H, Guan B, Cheng Q, Huang Z (2010) An Investigation on the Principal Paths to Plasma Oxidation of Propylene and NO. *Energy & Fuels* 24 (10):5418-5425. doi:10.1021/ef100451x

³ Penetrante BM, Brusasco RM, Merritt BT, Vogtlin GE (1999) Environmental applications of low-temperature plasmas. *Pure Appl Chem* Vol. 7 (No. 10):6



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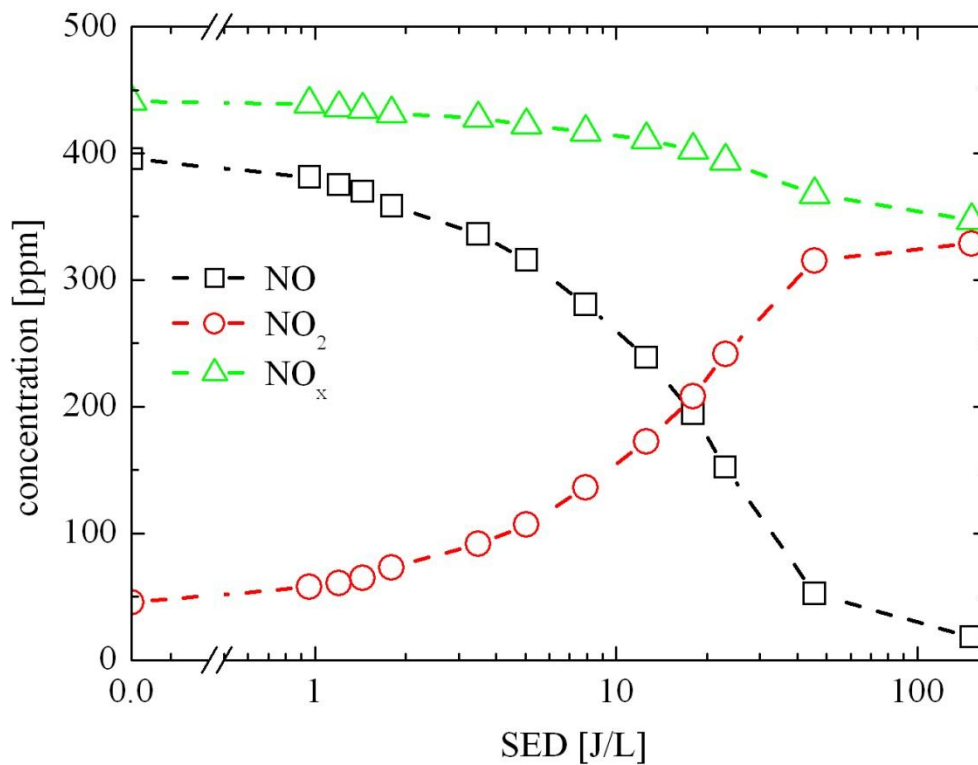
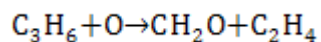


Figure 3: Concentrations of NO, NO₂ and NO_x with respect to the specific energy density

By-products

Due to the plasma activity some species are produced which are not added to the untreated gas mixture. These are referred to as by-products. The species known to be produced in gas mixtures like the one used in this experiment are N₂O, NH₃ and CH₂O. The concentrations of this by-products are given in Figure 4. The values of the N₂O-concentration (black boxes, left scale) are very small and the concentration of NH₃ (red circles, left scale) remains nearly close to zero. The only by-product produced in remarkable amount is CH₂O (green triangles, right scale). In^{2,4} the reaction



⁴ Guan B, Lin H, Cheng Q, Huang Z (2011) Removal of NO_x with Selective Catalytic Reduction Based on Nonthermal Plasma Preoxidation. *Industrial & Engineering Chemistry Research* 50 (9):5401-5413. doi:Doi 10.1021/ie1019744



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is given but due to the lack of ethylene this seems not to take place in this experiment. Therefore the reaction scheme including peroxy radicals given in⁵ is preferred to explain the chemistry.

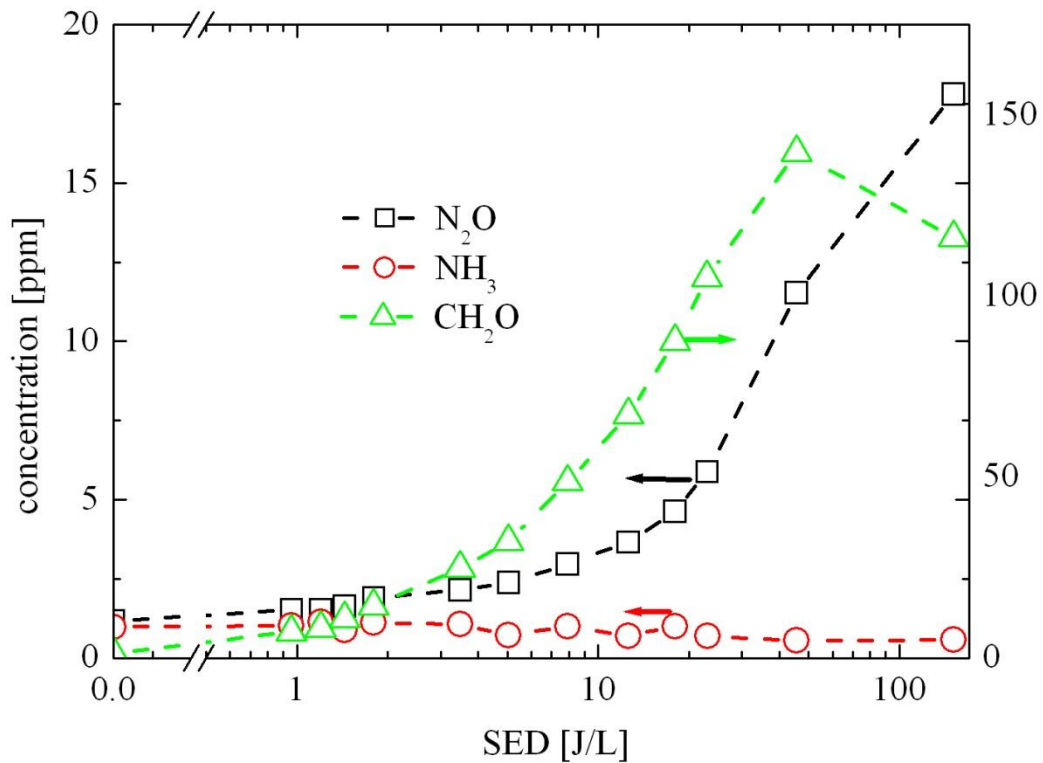


Figure 4: Concentrations of the byproducts with respect to the specific energy density

Water and carbon dioxide

For completion the concentrations of water and carbon dioxide are given in Figure 5. There are no noticeable changes in the concentrations in the range of per cent. In order to detect changes in the range of ppm water and carbon dioxide should be added to the background of the FTIR-measurement but this requires a significant change of the experimental setup and could not be realised up to now.

⁵ Martin AR, Shawcross JT, Whitehead JC (2004) Modelling of non-thermal plasma after treatment of exhaust gas streams. *J Phys D-Appl Phys* 37 (1):42-49. doi:10.1088/0022-3727/37/1/008



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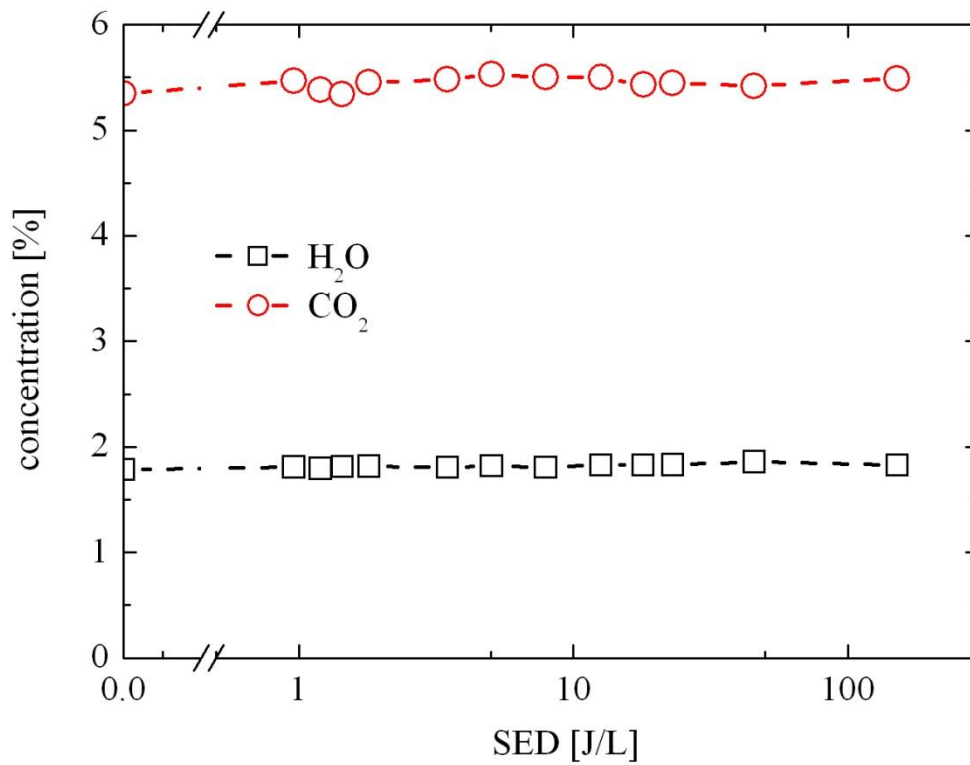


Figure 5: Concentrations of water and carbon dioxide with respect to the specific energy density

3. Summary and outlook

The main chemical pathways in the DBD stack-reactor were monitored as a function of the specific energy density. In a next step the energy yield for NO-conversion and NO_x-removal must be determined. However, the presented data already give the possibility to compare DBD treatment with other methods of plasma-supported exhaust gas cleaning, taking into account the specific conditions. Experiments with Toluene have been started.