Two-stage SDBD-reactor for VOC-treatment: Influence of the interstage gas residence time

M. Schmidt[1], M. Kettlitz[1], M. Schiorlin[1] and R. Brandenburg[1]

[1]*Leibniz Institute for Plasma Science and Technology, Felix-Hausdorff-Straße 2, 17489 Greifswald, Germany*

Abstract: A two-stage surface dielectric barrier discharge (SDBD) reactor is used to treat the volatile organic compound (VOC) toluene with an inlet concentration of around 300 ppm in dry air. The length of the connecting interface between the stages and thus, the gas residence time is varied systematically. It is found that increasing the interface gas residence time from 0.3 s to 7.6 s increases the conversion of toluene significantly from almost 30 % to around 70 % with the same plasma power of about 1.5 W. Also, the ratio of produced CO_2 to CO increases.

Keywords: surface DBD, multi-stage reactor, gas residence time, toluene removal

1. Introduction

The applicability of dielectric barrier discharges (DBDs) for cleaning of exhaust gases from volatile organic compounds (VOCs) is widely investigated. Operational parameters like the specific input energy, process gas humidity, the VOC to be treated, inlet concentrations, and reactor geometries were studied [1]. Applications, e.g. for cleaning of exhaust gases of professional kitchens, are already commercially available [2]. These systems utilize additional filters like active carbon or catalysts to improve the performance of the system. Another parameter influencing the chemical conversion of the pollutants is the gas residence time, meaning the time, which the polluted gas is in contact with chemical active species generated by plasma. The easiest way to vary this time is to set the size of the plasma source. This is, e.g. investigated in [3]. In this study, two subsequent corona discharges with variable length were used to remove toluene from air. It was found that the toluene removal increases with increasing reactor length and thus, the gas residence time. But, also the energy dissipated into the plasma increased. Varying the gas residence time by changing the flow rate did not lead to any significant difference in toluene conversion. In [4] three plasma sources connected in series for the removal of toluene from air was reported. Compared to one reactor, the use of three increased the toluene conversion by a factor of about 8 whereas the energy dissipated into the plasma increased by a factor of three, only. A similar experimental setup was utilized in [5]. Again, it was found that increasing the number of plasma stages and thus, the operational length of the plasma source increases the conversion efficiency. But, also the energy dissipated into the plasma increased with the length of the plasma source. To investigate the effect of residence time separately, we present an investigation with a two-stage plasma source with varying length but the power supplied to the plasma source was kept constant. The variation of the length was realized by varying the length of the connection pipes between the stages, and thus the gas residence time (interface time t_{IF}).

2. Experimental setup and proceeding

The experimental setup is shown in Fig. 1. It consisted of a surface DBD-reactor (SDBD), segmented in 5 channels. Only two of the channels were used to form the two-stage reactor. The connection between the channels was realized by pipes of varying length (30 cm, 280 cm, and 560 cm, respectively). The pipes were made of PFA with an inner diameter of 4 mm. Thus, the interface gas residence time t_{IF} was increased from 0.3 s to 7.6 s. The SDBD-reactor was energized with a programmable high-voltage source (Chroma 41604) and with a customized voltage transformer (Bremer). The power input was measured via the Q-V-plot method [6] with a measuring capacitance of 525 nF. The electrical data were analysed with a high voltage probe (Tektronix P6015A) and a 1:10 voltage divider (Tektronix P6139B) via an oscilloscope (Tektronix DPO4104). The gas supply was realized with gas cylinders (Air Liquide) and a bubbler for the admixture of toluene.

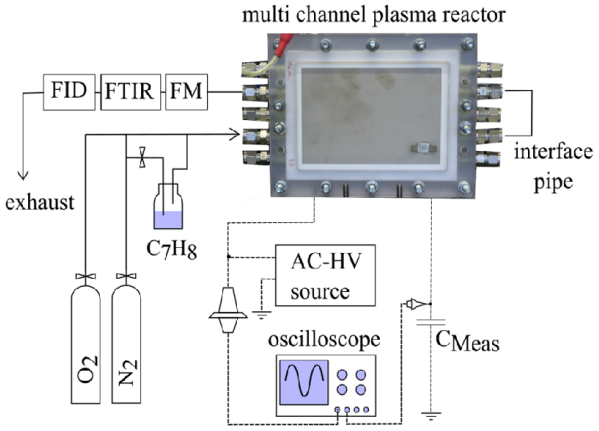

Fig. 1: Experimental setup

The gas flow was controlled by mass flow controllers (MKS Instruments). Gas analysis was performed with an FTIR-spectrometer (Bruker Alpha, T_{cell} = 40 °C, optical path length 5 m) and an FID (Testa 2010T). The total gas flow was measured with a flow meter (FM, Omega FMA 6610). The experiments were performed after stabilizing the toluene concentration at about 300 ppm. For the experiments, the interface pipes were chosen by increasing the length. After each measurement under "plasma on" conditions, the system was stabilized at the starting value. To ensure the repeatability of the experiments, this procedure was performed backwards (with decreasing interface pipe length) after measuring with the longest interface pipe. For all experiments the electrical parameter of the plasma source were kept constant (f = 400 Hz, U_{PP} = 14 kV, P = 1.5 W) to ensure the same power input.

3. Results and discussion

In Fig. 2 the removal of toluene from gas mixture of about 300 ppm toluene in synthetic dry air as a function of the interface time is shown. The toluene removal increases from around 30 % for t_{IF} = 0.3 s to 50 % for t_{IF} = 3.6 s up to ca. 70 % for t_{IF} = 7.2 s. These results were obtained just by varying the length of the interface pipe from 30 cm to 280 cm, and 560 cm and keeping the input power constant at 1.5 W. The investigated experimental setups are summarized in Table 1. The gas flow rate decreases with increasing interface length. This is most likely due to the increasing backpressure of the piping system. Accordingly, the specific input energy SIE calculated via equation (1), increases. In order to assure that

$$SIE\left[{}^{J}/_{L}\right] = \frac{input\ power\ [W]\cdot 3600s}{gas\ flow\ rate\ [{}^{L}/_{h}]} \qquad (1)$$

the higher toluene removal is not just an effect of the higher SIE, the power input into the plasma was varied with keeping the interface length L_{IF} constant (L_{IF} = 30 cm). It was found that for the investigated input power the removal

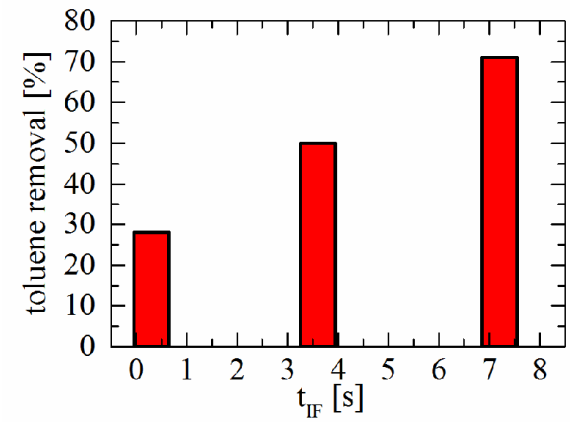

Fig. 2: Removal of toluene from gas mixture of about 300 ppm toluene in synthetic dry air for investigated interface times

Table 1: Experimental conditions and resulting parameters

interface pipe length	interface residence time	gas flow	SIE
cm	s	L/h	J/L
30	0.3	45	127
280	3.60	35	157
560	7.2	29	193

increased from 31 % to 41 %. This means, there is an effect of the input power but the main effect is due to the increased interface time t_{IF}.

Another effect of the varied interface time is shown in Fig. 3. Here, the concentrations of the reaction products CO_2 and CO are given. With increasing t_{IF}, thus with increasing removal of toluene, these concentrations also increase. According to [7] CO_2 and CO are the main oxidation products of the toluene removal process. Other reaction products found in this study were HNO_3 and CH_2O_2 as well as water, ozone, and N_2O which were not quantified because of lack of calibration spectra. It is remarkable that the ratio of CO_2 to CO increases with increasing t_{IF}. Obviously, the higher residence time lead to more oxidizing reactions, which favours CO_2-selectivity.

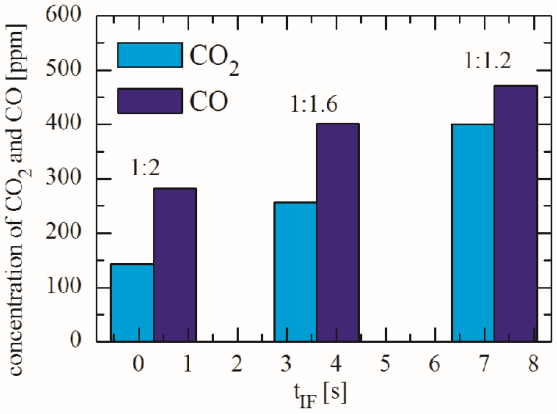

Fig. 3: Concentration of CO_2 and CO for varying t_{IF}, the numbers on top of the bars give the according CO_2-to-CO-ratio

4. Summary

In this contribution, a two-stage SDBD-reactor is utilized for removal of about 300 ppm toluene from synthetic dry air. To investigate the influence of the gas residence time on the removal process the length of the gas pipe between the two stages was varied and thus, the gas residence time (interface time t_{IF}) whereas the power input into the plasma was kept constant. With increasing t_{IF} not only the toluene removal increased but also the CO_2-to-CO-ratio, showing that more oxidizing reactions take place when the residence time is increased.

5. References

[1] A.M. Vandenbroucke, J. Hazard. Mater., **195** (2011).
[2] https://www.baero.com/de/clean-air-technologies/ (in German).
[3] R. McAdams, Plasma Sources Sci. Technol., **16**, (2007).
[4] A. M. Harling et al, Ind. Eng. Chem. Res., **47** (2008).
[5] M. Huebner et al, Journal of Applied Physics, **114**, (2013).
[6] T.C. Manley, Trans. Electrochem. Soc., **84** (1943).
[7] M. Schiorlin et al., Environ. Sci. Technol., **43**, (2009),