

DBD PLASMA PROCESS FOR EXHAUST AIR PURIFICATION

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1 Introduction

For the application of exhaust gas treatment the NT- plasma technology in combination with catalysts has big potential especially for odor reduction and the reduction of VOC's at lower concentration (up to $500 \text{ mg}\cdot\text{m}^{-3}$) in industrial production plants. For basic investigation a DBD plasma module especially constructed for exhaust gas treatment, in combination with a low-cost catalyst is investigated. The NT- plasma module is suited to be operated directly in the exhaust gas stream or in bypass mode and capable for treatment of $500 - 1500 \text{ m}^3\cdot\text{h}^{-1}$ of exhaust gas. By switching modules in parallel, the units are suited to be operated for very big volume flows. Basic investigation about the plasma composition and the decomposition potential were conducted. Ammonia and n-butanol are used as model contaminants.

2 Experimental setup

The setup for the waste gas treatment consists of a dielectric barrier discharge (DBD) reactor in combination with a low cost mineral adsorber. Fig. 1 shows a schematic view of the experimental setup.

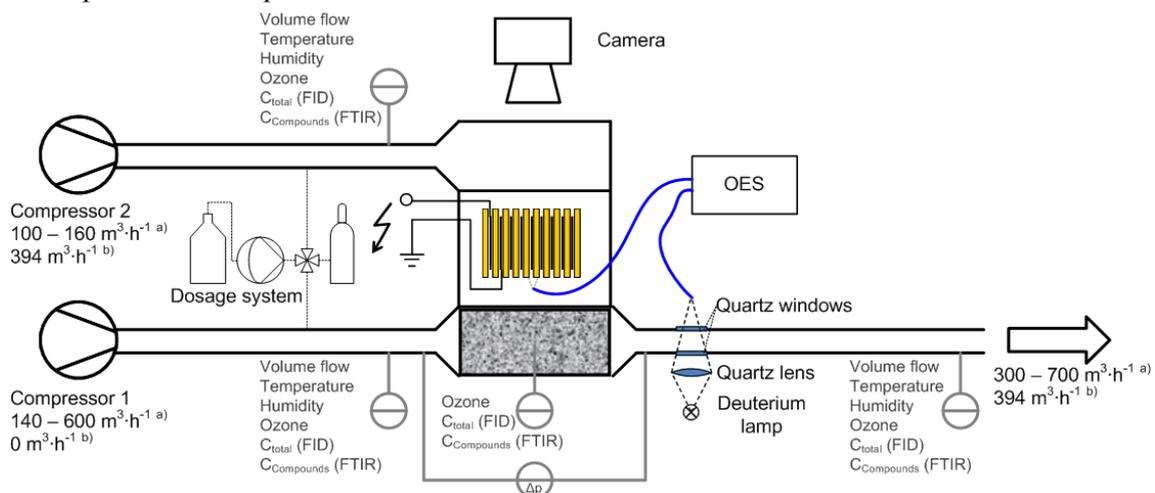


Figure 1: Scheme of the experimental setup. ^{a)} Volume flows for indirect mode, ^{b)} volumes flow for direct mode.

The whole system was used for gas volume flow rates of $394 \text{ m}^3 \cdot \text{h}^{-1}$ in direct treatment mode and, additionally, $300 \text{ m}^3 \cdot \text{h}^{-1}$ to $700 \text{ m}^3 \cdot \text{h}^{-1}$ in indirect mode. To generate a defined synthetic waste gas noxious gas content can be added up in defined concentrations. In case of the direct mode, the waste gas is fed directly into the DBD. The indirect mode is used in case of corrosive gases. Here, neutral air passes through the DBD, is activated and subsequently mixed with the waste gas. An adsorber material is placed further downstream to increase the residence time, thereby increasing the degradation of contaminants. The adsorber material preferably consists of low-cost materials like bentonite (GA1) and halloysite (GA2).

The DBD is constructed as a stack of 16 powered and 17 grounded electrodes separated by 34 ceramic barrier plates. At both ends a grounded electrode with a ceramic barrier terminates the stack. The system is powered by a low frequency high voltage mains supply. The frequency ranges from 320 Hz up to 720 Hz at a maximum power of 3 kW. The applied power is adjusted by the discharge current, which in turn defines the discharge voltage.

To characterize the DBD plasma, the plasma homogeneity is observed by a camera system via a telescopic configuration from a distance of 25 m. This enables a very narrow observation angle and therefore the possibility to see into all the discharge gaps of the DBD stack. The shape of the plasma discharge is observed by a close-up view and is characterized by optical emission spectroscopy (OES). The OES spectrometer is a combination of 3 CCD spectrometers and comprises the wavelength range from UV light up to NIR light. The plasma emission is collected by a spectroscopic fiber system. For the detection and data logging of the produced ozone (O_3), the OES spectrometer is also shared for the O_3 light absorption measurement of UV light at 254 nm.

To correlate the plasma homogeneity with the treatment efficiency of the system n-butanol and ammonia were added as sole contaminants. For identification and quantification of both contaminants as well as their secondary reaction products after exposure to the plasma, a FID and a FTIR were used. In addition, the use of adsorbers was compared to the process without adsorber due to transformation efficiency as well as minimization of critical reaction products like N_2O .

3 Experimental results

The DBD reactor was visually characterized as explained in the previous section, and it was noted that the overall discharge looks more homogeneous towards lower frequencies and higher applied powers. This effect is pictorially represented in fig. 2.

The input power chosen by the primary current of the transformer was varied from 0.7 kW up to 2 kW. At a high power and low frequency the generator had to increase the output voltage to ignite enough micro discharges in a half-wave. This led to an excellent homogeneity. At certain values the high voltage led to an arc generation and the arc detector of the power generator shut down. Due to the arcs no photos of the discharge at critical parameter sets can be presented in fig. 2.

The OES investigation via fiber spectrometer revealed astonishingly a dull light emission. The only lights which can be detected were the molecular emission bands of the N_2 second positive system $C^3\Pi_u-B^3\Pi_g$. They occurred in the wavelength range from 290 nm to 440 nm. The visible light emission shown in fig. 2 originated from the violet-blue part of the N_2 band.

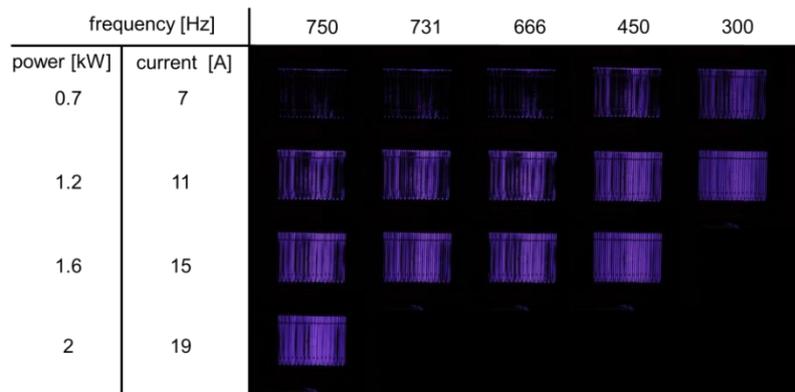


Figure 2: Photographs of the plasma homogeneity for frequencies of 300 Hz to 750 Hz and the power from 0.7 kW to 2 kW.

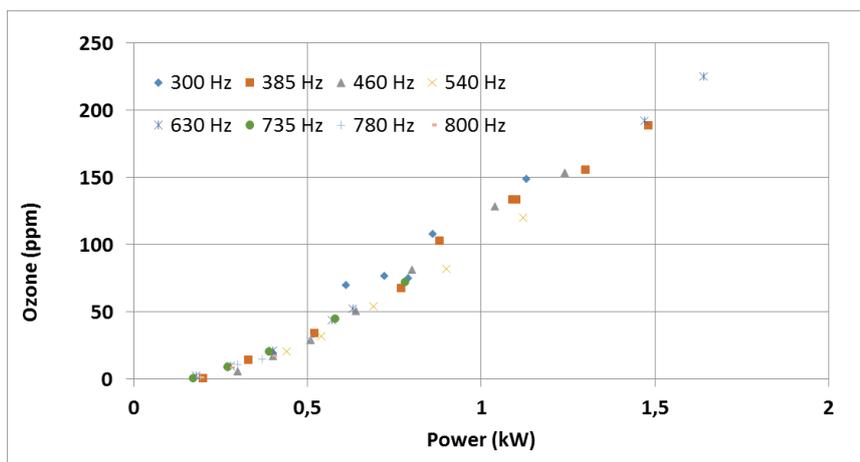
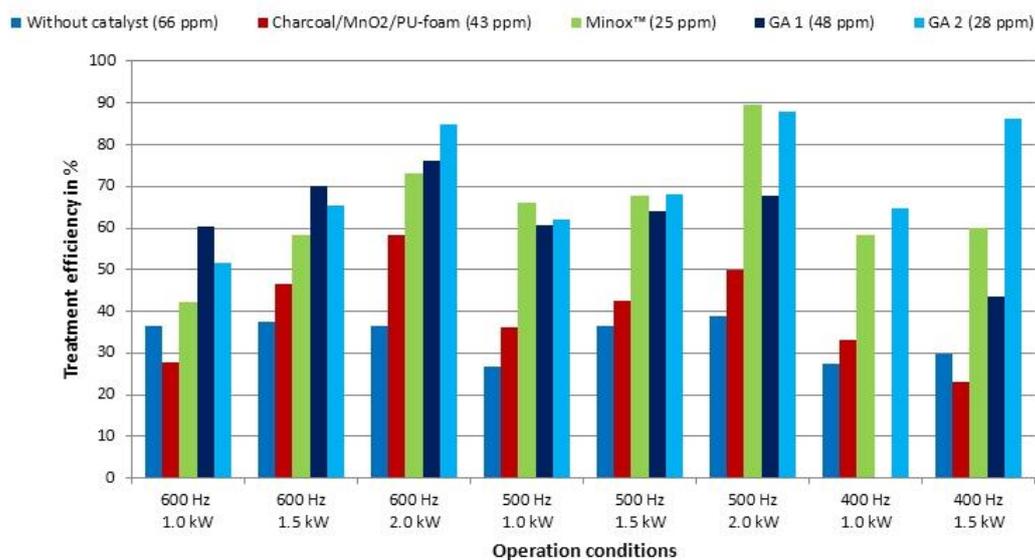


Figure 3: Ozone generated vs. applied power. The ozone generation is solely a function of the applied power.

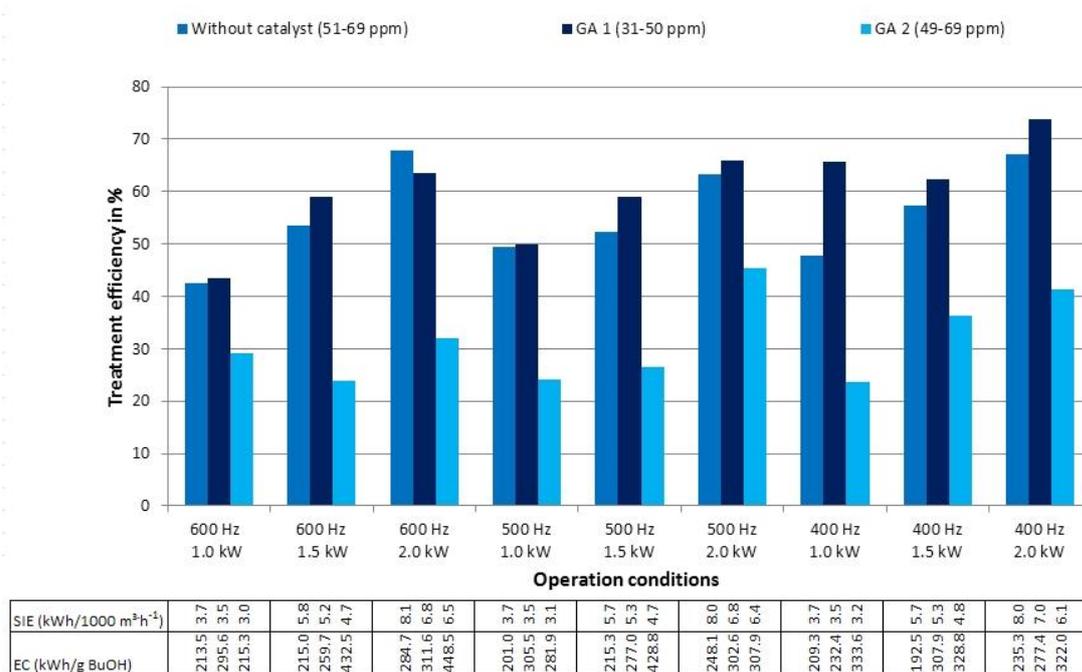
In a similar manner as visual observation, the frequency and current were varied and the ozone produced was measured in the outlet duct of the reactor using UV absorption spectroscopy at 254 nm. The Beer-Lambert law [1] was used to calculate the ozone concentrations.

It was observed that the ozone generated was solely a function of the applied power at a given air flow ($200 \text{ m}^3\text{h}^{-1}$ in this case for conservative estimates), as can be seen in figure 3. Further investigations are needed to clarify the exact reaction paths for the production of ozone and activated species and their reaction mechanisms with the pollutants.



SIE (kWh/1000 m ³ h ⁻¹)	41.8	3.3	5.0	6.7	3.3	5.0	6.7	3.3	5.0
EC (kWh/g BuOH)	84.9	95.6	34.8	69.6	61.2	75.7	103.8	44.9	82.4
	83.6	80.5	110.4	55.1	85.0	57.3	65.0	61.0	34.7
	58.1	62.7	82.8	89.2	49.1	79.4	78.7	94.2	90.0
	62.1	81.9	55.4	70.5	69.2	n.a.	55.5	77.1	153.7
	100.8	72.5	62.7						

Figure 4: Treatment efficiency for n-butanol of the combined plasma-adsorber system vs. input power in indirect mode with a gas volume flow of 300 m³·h⁻¹.



SIE (kWh/1000 m ³ h ⁻¹)	213.5	3.7	3.5	215.3	3.0	215.0	5.8	284.7	8.1	201.0	3.7	215.3	5.7	248.1	8.0	209.3	3.7	192.5	5.7	235.3	8.0
EC (kWh/g BuOH)	295.6	215.3	215.0	259.7	432.5	448.5	311.6	305.5	281.9	277.0	428.8	302.6	307.9	232.4	333.6	307.9	328.8	307.9	328.8	277.4	322.0
	3.0	4.7	6.5	5.2	6.5	6.5	6.8	3.5	3.1	5.3	4.7	6.8	6.4	3.2	4.8	4.8	4.8	5.3	4.8	7.0	6.1

Figure 5: Treatment efficiency for ammonia of the combined plasma-adsorber system vs. input power in indirect mode with a gas volume flow of 300 m³·h⁻¹.

To improve transformation rates of the plasma stage for both contaminants as well as secondary intermediates, high specific loading capacities of the adsorber material are required. Thus, adsorption isotherms of three bentonites and two halloysites were

performed in the range of 0 – 325 mg n-butanol·m⁻³ and 0 – 475 mg NH₃·m⁻³, showing a linear behavior between concentration and specific loading capacity of the adsorber. With restriction to commercial availability the bentonite GA1 and the halloysite GA2 were chosen for further application, even though GA2 did not show highest performance. The specific loading capacities of both materials were 4.86 w% and 2.15 w% (GA1) as well as 2.60 w% and 1.77 w% (GA2) in case of 100 mg C·m⁻³ of n-butanol or 100 mg N·m⁻³ of ammonia, respectively. These values are similar to those of charcoal systems.

Further studies about the transformation rates of the combined plasma – adsorber system revealed a direct correlation between the specific loading capacity of the adsorber and the overall treatment efficiency of the system. The ratio of the loading capacities of GA1 to GA2 in case of 100 mg n-butanol·m⁻³ was 1.86, which is acceptably corresponding with the inverse ratio of the specific energy consumption (EC) of 1.66 in average (see figure 4). In case of ammonia a perfect fitting of this correlation occurred with values of 1.21 in case of loading capacities and 1.22 in case of EC (see figure 5). For n-butanol similar effects were even observed comparing both clay materials with charcoal. In general, both increasing specific input energy (SIE) as well as a reduction of input freight of contaminants helped to improve the efficiency of the system significantly. The effect of SIE on the formation of critical secondary metabolites like N₂O or NO was contrarily low. Only NO₂ and butanal accumulated as main products of the conversion of ammonia and n-butanol. Finally, the presence of adsorbers helped to improve the n-butanol conversion efficiency significantly, but showed indifferent effects in case of ammonia.

Summary

Within this project experimental setups were build that allows optical and DRE minvestigations for VOC treatment of the NT- plasma unit with simulated and real exhaust gases in a technical scale.

It was found that the plasma generation has a good homogeneity at the foreseen operation point and the ozone generation follows a linear correlation with the input power.

Treatment efficiencies were measured at different plasma input power and concentrations. The presence of an adsorber helped to improve the n-butanol conversion efficiency of the system significantly, but showed indifferent effects in case of ammonia.

Investigations revealed a bundle of optimization approaches, which where implemented in a pilot scale system further qualified with real exhaust gases (i.e. digestion tower, bio-mechanical treatment, sludge drying).

Acknowledgements

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References

- [1] W.B. DeMore, M. Patapoff, *Environmental Science and Technology*, 10(9):897 (1976)