Scale-up of a coaxial DBD reactor for the oxygen removal from coke oven gas with non-thermal plasma

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Abstract: Trace oxygen removal from coke oven gas is desired for more economical hydrogen recovery by pressure swing adsorption. A non-thermal plasma in a coaxial DBD reactor removes 90% of 1000 ppmV oxygen in coke oven gas at a specific power input of 1,440 J/L. By maintaining this specific power input, the same removal outcome is achieved for volume flow rates up to 0.3 Nm³/h. Further modifications of the reactor geometry promise to further reduce the specific power input and thus the process costs.

Keywords: non-thermal plasma, coke oven gas, scale-up, oxygen removal, gas conditioning

1. Introduction

The research project Carbon2Chem® aims to use CO_2 from steel mill gases to produce chemical products such as methanol or urea [1]. The key compound for the conversion of CO_2 into value-added products is hydrogen. Furthermore, conditioning of the steel mill gases is essential to enable optimal reaction conditions for the desired syntheses. Thus, the treatment of the coke oven gas (COG) plays an important role as it contains up to 65 vol% hydrogen. For a flexible and economic conversion, the hydrogen must be recovered from the COG. One method therefore is the pressure swing adsorption (PSA).

However, COG contains low concentrations of oxygen. This oxygen in the COG comes from, for example, air leaks in the coke plants or chemically bound oxygen in the coal. Oxygen is an impurity for the PSA that must be removed, but more importantly, explosive mixtures can occur during the PSA process under certain conditions. Therefore, an upstream removal of oxygen is desired. Catalytic or adsorptive oxygen removal is very limited for this application due to the high variety of trace components in the COG such as ammonia, sulfur or organic compounds.

Non-thermal plasma is considered to be less affected by catalytic poisons and is therefore a potential alternative for the oxygen removal. Laboratory experiments have already shown that non-thermal plasma can remove oxygen in a model COG without relevant changes in the main components [1]. In a laboratory glass reactor, up to 90 % of oxygen could be removed with non-thermal plasma at a total volume flow rate of 0.1 Nm³/h and a power input of 40 W (specific energy input SEI: 1,440 J/L).

However, one ton of steel produces already 50 Nm³ of COG [3]. Thus, the technology needs to be scaled up. While coaxial Dielectric Barrier Discharge (DBD) reactor systems can be already numbered up successfully as shown for ozone generation systems [4], the scale-up potential of this system has not fully investigated yet and is a potential to minimize the reactor size and cost at a given volume flow rate.

2. Experimental Setup

The test system provides gas mixtures for typical steel mill gas compositions (main components: H_2, CH_4, CO_2, CO, N_2; exemplary minor components: O_2, H_2S, toluene, propane). The plasma reactor (Fig. 1) is based on the annular/packed-bed DBD design and is designed with a steel jacket and an inner glass tube as the dielectric [3]. The reactor is designed to handle volume flow rates up to 1 Nm³/h with gas hourly space velocity of 3,600 h^{-1}.

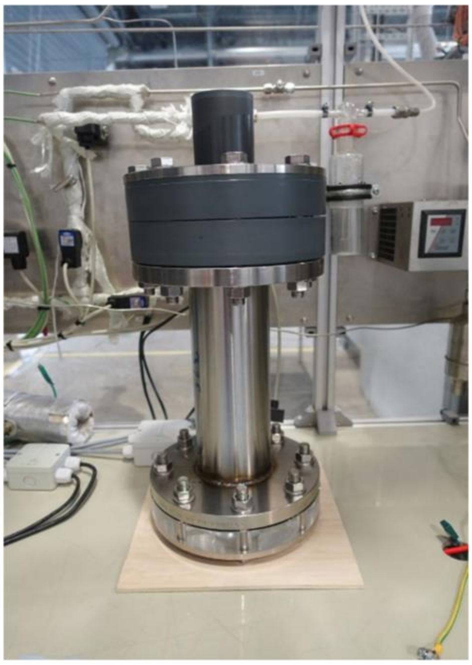

Fig. 1. Photography of the investigated coaxial DBD reactor system

The reaction volume is set by the length of the discharge region. The plasma is generated at gap widths between 2–4 mm. The high-voltage generator provides the reactor with voltages up to 20 kV_{pp} in a LC resonance circuit. This circuit can be amplified with voltage pulses up to 300 V at frequencies from 4–500 kHz. An additional duty cycle in the ms-range limits the power input as well.

3. Results

The steel reactor allows a reproduceable high conversion of oxygen over 90 % with 40 W at 0.1 Nm^3/h in comparison to the previous investigated glass reactor [2]. However, increasing volume flow rates and oxygen inlet concentrations are decreasing the conversion at the same power input. This is observed at DoE screening experiments (Fig. 2).

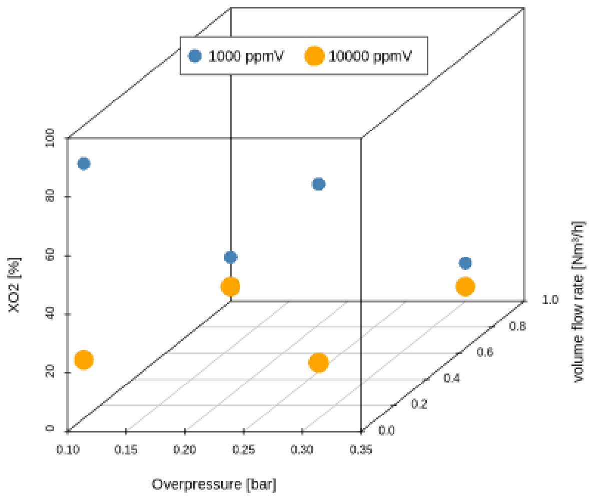

Fig. 2. Screening of oxygen removal in a COG at 40 W in the coaxial DBD reactor with 2 mm discharge gap

The conclusions of this results clearly indicate that with a power input of 40 W the transferred energy into the electrons is not high enough to initiate the conversion of a higher amount of oxygen molecules. For this reason, the proportional increase in power consumption is examined (Fig. 3).

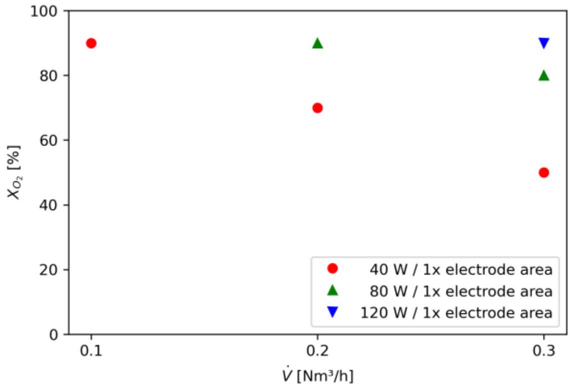

Fig. 3. Screening of oxygen removal rate in a COG at 40W with a steel DBD reactor with 2 mm discharge gap

A continuous power input allows to upscale the oxygen removal process. However, the SEI of 1,440 J/L is quite high compared to other gas cleaning processes as the oxidation of volatile organic compounds (VOC) with 100–250 J/L [5]. Thus, further adaptations of the reactor are investigated as the reactor geometry and the implementation of catalysts. Furthermore, the electrical parameters of the high voltage generator are varied in order to increase the efficiency. The examined reactor modifications as well as the results are shown at the symposium.

4. Conclusions

Increasing the oxygen inlet concentration and the total volume flow rate of a non-thermal plasma oxygen removal process at constant power consumption results in a decrease in conversion. A constant specific energy input of 1,440 J/L allows the conversion result to be maintained at higher volume flow rates. However, the absolute value of the SEI is high. Therefore, further geometrical variations of the scaled up reactor as well as electrical parameters are investigated.

5. Acknowledgements

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6. References

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