Plasma-catalytic removal of oxygen traces in steel mill gases with a packed-bed DBD reactor

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Abstract: The conversion of oxygen traces (500–1500 ppmV) in synthetically generated coke oven gas mixtures with non-thermal plasma-catalysis is investigated. Non-thermal plasma converts trace oxygen in coke oven gas mixtures without catalyst at conversion rates of 20–30 % with 15–20 W power input up to now. The combination of non-thermal plasma with Pt/Al_2O_3-catalyst or Al_2O_3-granules shows in first tests improvement of the conversion as well as the variation of the input plasma parameters and the residence time.

Keywords: non-thermal plasma, plasma-catalysis, steel mill gases, plasma reactor

1. Introduction

The research project Carbon2Chem® aims at utilizing CO_2 from steel mill gases for the production of chemical products (e.g. methanol).As a consequence, the CO_2-emissions of the steel mill and the use of fossil fuels in the chemical industry can be reduced[1]. An essential process step for the conversion of the steel mill gases is the conditioning of the exhaust gases. One specific challenge is the selective removal of trace components. Therefore, chemical conversion processes are preferred.

Catalysts are used for industrial chemical conversions to reduce the required process temperature as well as to adjust the selectivity of the reaction network. Opportunities for further improvement of the conversion are the external excitation of reactants or active centers of the catalyst. The excitation allows even further reduction of the reaction temperature in certain reaction setups. Microwaves [2], radiation of UV or visible light [3] and plasma [4] are well known technologies for external excitation.

The potential of non-thermal plasma in combination with a catalyst (non-thermal plasma-catalysis) is already intensively investigated for removal of trace components such as volatile organic compounds (VOC) in air [1] and NO_x in diesel exhaust gases [2]. Within the research project Carbon2Chem® the potential of non-thermal plasma-catalysis is investigated for the removal of trace oxygen from the coke oven gas of the steel production. The oxygen traces in the coke oven gas are the result of air leakage at technical plants and exhaust pipelines and can influence subsequent conditioning and synthesis steps. As a consequence, the oxygen traces have to be selectively removed to protect downstream process steps (e.g. PSA).

General feasibility of the process with respect to industrial scalability of plasma-catalytic reactor systems must be proven. The focus of the experimental program is examining the non-thermal plasma-catalytic conversion of these oxygen traces. For this purpose, an annular/packed-bed DBD plasma reactor has been assembled at Fraunhofer UMSICHT (Chapter 2), while a surface-DBD setup is tested at Ruhr-University Bochum [5].

2. Experimental Setup

A test rig for the plasma-catalytic conversion has been designed and assembled. The test system provides gas mixtures for typical steel mill gas compositions (main components: H_2, CH_4, CO_2, CO, N_2; minor components: O_2). The plasma reactor (Fig. 1) is based on the annular/packed-bed DBD design [3].

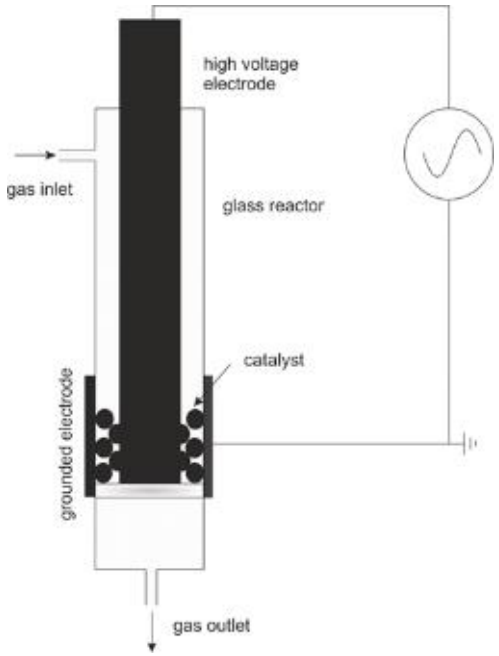

Fig. 1. Setup of the plasma-catalytic reactor (annular/packed-bed DBD design)

The reactor allows conversions in the residence time range of 1–2 s (= GHSV of 1800–3600 h^{-1}). The reaction volume is defined by the length of the discharge region. The annular discharge gap of the packed-bed-DBD tested at 2 and 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. Additionally, a duty cycle can limit the power input.

3. Results

The usage of non-thermal plasma allows the conversion of oxygen traces with oxidizable components of the coke oven gas as H_2, CH_4 and CO, Applying non-thermal plasma, oxygen conversion rates of 20–30% are achieved with a power consumption of 15–20 W. If the discharge gap is increased to 4 mm, a power input of 35 W is necessary for the same conversion outcome. The oxygen conversion can be increased by variation of the voltage, which influences the power input (Fig. 2).

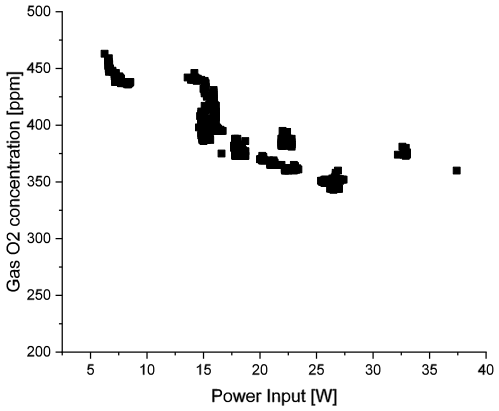

Fig. 2. . Resulting O2-conversion in annular DBD plasma reactor with 2 mm discharge gap

The selected DeOxo-catalyst system (Pt on Al_2O_3) allows complete conversion of O_2 traces with hydrogen, which is the main component of the coke oven gas (up to 60 %). Unfortunately, the catalyst is deactivated by CO, which is present with 5-7 Vol% in the coke oven gas mixture as well. As a result, the catalyst is not converting trace oxygen in coke oven gas at ambient conditions In contrast, non-thermal plasma allows partial O_2 conversion in presence of CO.

In preliminary tests it could be shown, that the placement of catalyst granules in the discharge zone (in-plasma-catalysis) allows improvement of the oxygen conversion compared to the non-thermal plasma treatment. In ongoing tests it has to be investigated, if the platinum catalyst or the Al_2O_3-granules itself improve the conversation rates at ambient temperatures. First results of these investigations are very promising for the optimization of the oxygen conversion.

4. Conclusions

While the feasibility has been clearly demonstrated, the conversion rate has to be further optimized by geometrical adaption of the reactor setup. This has to be done to establish a homogeneous and easier controllable plasma treatment. Afterwards the next scale-up step towards a pilot installation using real process gas streams can be taken. More detailed results will be presented at the symposium.

5. Acknowledgements

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6. References

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