The use of a nanosecond-pulsed corona plasma for tar-cracking at high temperatures: first insights

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Abstract: Tar is the toughest barrier that hampers the use of syngas in top-notch applications. Cold plasmas have being successfully used for tar removal at temperatures below 400°C, but for energy efficiency purposes, tar cracking should be done beyond 600°C. In this paper we present the effect of a nanosecond-pulsed corona discharge over naphthalene cracking at 600°C on a N_2 atmosphere. The results show a drastic reduction of naphthalene concentrations when compared with thermal cracking at the same temperature.

Keywords: nanosecond plasma, pulsed corona plasma, syngas cleaning, tar removal , naphthalene cracking.

1. Introduction

Dielectric barrier discharge (DBD) plasma [1-3] and DBD plasma-enhanced catalytic systems [4-5] have been used successfully for the removal of tar model molecules at temperatures ranging from 25°C to 400°C . Since exit temperatures of gasifiers range from 600°C up to more than 1200°C, there is a need of operating such cold-plasma cracking units at temperatures above 400°C. This threshold is difficult to overcome for current DBD plasma units because of the thermal tolerance of the materials utilized, especially as dielectric material, which include plastics, silicon rubber and Teflon [6]. Some other materials in DBD such glass, quartz, ceramics, enamel or mica can stand high temperatures, but limit the peak voltage of the resulting plasma.

The use of a tar cleaning method at temperatures above 600°C is justified by the temperature of the syngas exiting traditional gasifiers. In fixed-bed gasifiers gas exit temperature ranges from 450 to 650°C, in fluidized beds 800–1000 °C and in entrained-flow the temperatures are above 1200°C [7].

To tackle this challenge we present a lab-scale direct-current (DC) nanosecond-pulsed corona plasma unit able to operate at temperatures as high as 1200°C. The discharge characterization of the plasma shows us that the pulse repetition is around 90 pulses per second with a pulse duration of hundreds of nanoseconds.

2. Experimental setup

The experimental setup consisted of a Kanthal tube (an alloy of FeCrAl) of 1.5 meters with a diameter of 64mm embedded in a three zone oven. Coaxial to this tube there is a Wolfram wire that is connected to a high-voltage (HV) circuit. The naphthalene is fed to the reactor by means of a saturator working with a precision of ±1°C, through which a stream of nitrogen passes by. The saturated nitrogen stream can be mixed with a pure nitrogen stream in order to keep the naphthalene concentration constant.

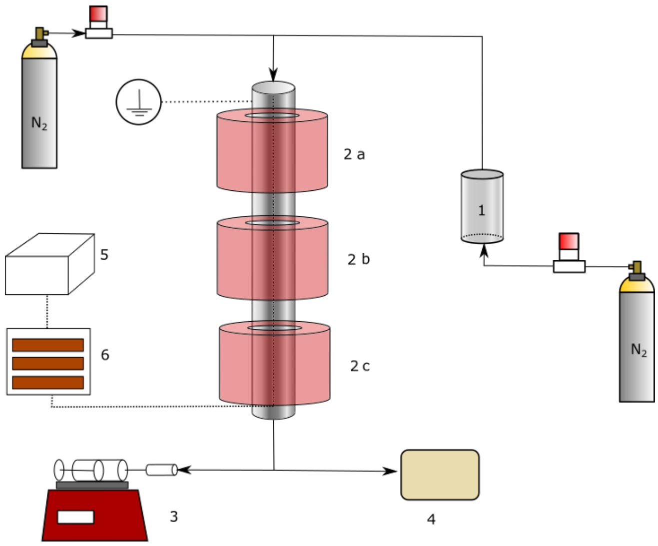

Fig. 1. Corona Plasma setup. **1** Saturator, **2a.2b.2c**. Three zone reactor temperatures, **3.** SPA/SPE sampling, **4.** Exhaust, **5.** High-voltage generator, **6.** Pulse generator. Dotted lines represent electric connections, solid lines represent gas connections.

The HV circuit consists of a DC-HV power supply connected to pulse generator, which consisted of an RC circuit using an spark gap to regulate the pulses generated. 90 pulses per second in average were generated throughout all the experiments. The temperature in the three zones of the reactor was kept at 600°C with deviations of ±40 °C from wall to wall.

The naphthalene was measured by the SPA/SPE method. An SPA column was placed at the exit of the reactor and 100ml of gas were sampled by using a syringe pump. The pressure of the gas was measured by a barometer to evidence any parasitic air dilution. The SPA column was flushed with 2ml of Dichloromethane (DCM) before sampling, and with 2ml of DCM after sampling to extract the adsorbed naphthalene. The extracted liquid was analysed using a GC-MS.

The initial concentration of naphthalene was determined at 110°C before each experiment, and between each experiment a stream of clean nitrogen was fed in order to have a clean reactor for the upcoming experiment. All the lines downstream the saturator were heated at 105°C including the SPA sampling lines.

The plasma pulses were measured by a Handyscope HS26 from the company TiePie which allowed to measure the voltage and current intensity.

3. Results and discussion

The plasma discharge can be in figure 2. At 600°C the plasma showed an average of 90 pps, The HV source was set at 24 kV resulting in a peak voltage of around 28kV and in a peak current of 250 A. The peak voltage duration is around ~30ns. The voltage curve shows an oscillating behaviour with two other peaks of the similar duration but with less voltage. The current showed one peak which lasted around 120 ns. The resulting average energy was 140 mJ / pulse.

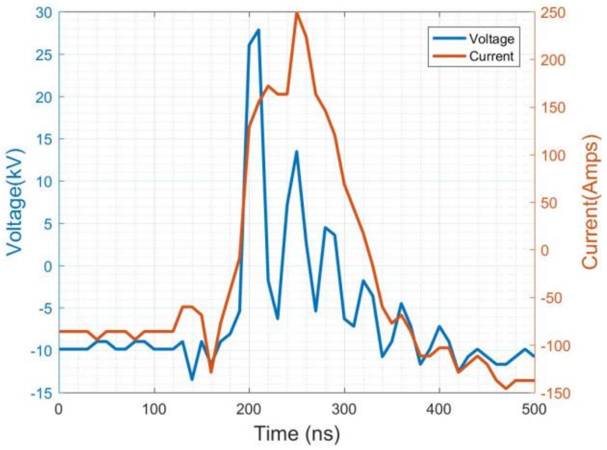

Fig. 2. An oscilloscope view of voltage (blue) and current (orange) of a nanosecond pulse in the corona plasma. Peak voltage around 26kV with a duration of ~30ns is observed.

The initial concentration of naphthalene was kept by maintaining the saturator at 70±1°C. The residence time was kept around 4 minutes, and the temperature of the reactor at 600°C. In these conditions, 4 different samples of naphthalene were taken with and without the pulsed corona plasma. The results are shown in figure 3.

The enhancing effect of the plasma is evident. At each case the corona plasma achieved much lower concentrations of naphthalene than the thermal conversion alone. However the effect does not seem to be equal for all the samples. This could be a result of the inhomogeneous nature of the plasma, a short number of samples taken by experiment or even a catalytic effect of the FeCrAl oxide in the reactor walls.

The corona plasma reactor show also potential for long-term operation, since the pulses current and voltage characteristics remained constant throughout all the experiments, which can be an advantage with respect to catalysts, or can be also a way of improving catalytic tar removal for long-run purposes.

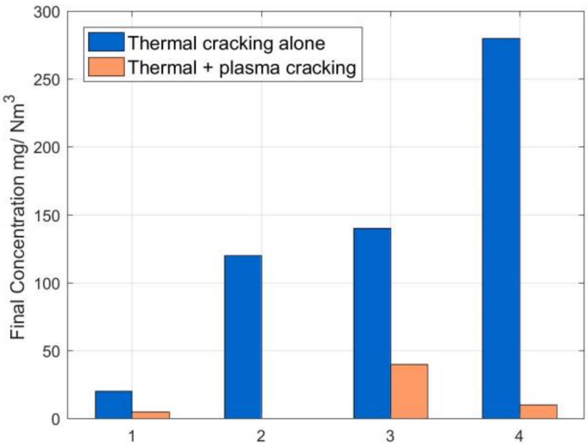

Fig 3. Naphthalene concentrations in N_2 at 600°C in the three zones of the reactor in the presence of plasma (orange) and in the absence of plasma (blue). The experiment is repeated 4 times with the same conditions.

4. Conclusions

After a first set of experiments, where a nitrogen stream with different concentrations of naphthalene is subjected to a temperature of 600°C in the presence and in the absence of plasma, the results show a clear cracking enhancing effect when pulsed-corona plasma is applied. The use of DC pulsed-corona plasma is thus promising for tar abatement in syngas, transforming syngas from biomass or waste gasification to a valuable product for further energy and/or material valorisation.

Acknowledgements

The research leading to these results has received funding from the European Community's Horizon 2020 Programme under Grant Agreement No. 721185 (MSCA- ETN NEW-MINE).

5. References

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