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Commissioning of a pulsed power corona discharge reactor for CO_2 reduction

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Abstract: The design and commissioning of a pulsed power corona discharge reactor for CO_2 reduction has been completed. The wire-in-tube reactor has been successfully paired with a high voltage power supply capable of producing 10 ns pulses up to 40 kV in magnitude. During the commissioning process a number of test experiments were completed. During this time, proof that CO_2 could successfully be reduced to CO was achieved and the best conversion observed was 14.08% (9% CO_2 mixture in Argon, 100 ml min^{-1}, 6 kV, 2000 Hz).

Keywords: carbon dioxide, CCU, corona discharge, pulsed power

1. Introduction

This research is undertaken as part of 4CU, a flagship ESPRC funded project for Carbon Dioxide Capture and Utilisation (CCU). The aim is to produce economically viable materials from a carbon dioxide feedstock, the main goal being to create synthetic fuels either via a Fischer-Tropsch type process or by direct catalysis. The reaction pathway involves carbon dioxide (CO_2) reduction to a carbon monoxide (CO) intermediate, followed by reaction of carbon monoxide to produce a useful product.

The work reported is part of a sub-project within 4CU focusing on carbon monoxide production. Non-thermal plasmas, specifically pulsed corona discharges are used as they already have proven industrial-scale application in electrostatic precipitators to remove particulates from waste gas streams in, for example power stations and cement factories. The utilisation of high voltage electricity passed through wires or points with a narrow curvature generates a high local electric field from which a corona discharge is established. The objective is to take recognised corona discharge technology and apply it in novel way. Carbon dioxide gas is passed through a strong electrical field wherein electron excitation occurs so that the gaseous feed becomes partially ionised and, upon recombination, a fraction of carbon monoxide is formed along with ozone and oxygen as minor products.

Using a combination of numerical and experimental techniques, the aim of this work is to develop a laboratory-scale plasma system that has the potential to be scaled to industrial levels capable of contributing towards the reduction in global carbon dioxide levels and provide an economic and efficient method of CO2 utilisation.

2. Experimental Set Up

The reactor is of stainless steel tubular design with an interchangeable wire electrode running centrally down the tube (for commissioning this was made of copper). Polyether ether ketone (PEEK) made flanges at either end of the reactor seal the vessel and serve as the connection point for gas inlets and power lines. Important dimensions for the reactor are the volume, 300 ml, and the inter-electrode distance, 15 mm.

The required power supply must be capable of producing high voltage pulsed DC electricity. Experience with industrial electrostatic precipitators has shown that pulsed power is the most energy efficient method to generate a plasma and enhances the electrical properties of the plasma allowing stronger electric fields to be generated. The short duration of the applied pulses prevents arc-type discharges from developing in the reactor as there is not enough time for an arc to travel between electrodes [1].

The power supply (Megimpulse Ltd.) is capable of producing high voltage (HV) pulses up to 40 kV, 10 ns in duration with a frequency up to 3.2 kHz. Voltage is measured with PVM-3 HV probe (Northstar), current by a Pearson 6595 current monitor and the data is transferred to a computer via a Picoscope 6404c oscilloscope.

3. Electromagnetic Inference and X-rays

An issue found during commissioning was electromagnetic radiation generation from the reactor and power supply system in such large quantities that it caused severe interference with surrounding electronic equipment. It was observed that using an electrode with a lower work function worsened the problem of electromagnetic interference (EMI). A lower work function produces more electrons and therefore a plasma can be ignited more easily i.e. the resistance of the gas is decreased. This allows more current to flow throughout the plasma and thus, the amount of EMI is increased.

This phenomenon occurs due to the fact the power supply produces high voltage pulses at a high repetition rate, a source of electromagnetic radiation [2]. The internal wire electrode acts as an amplifier to electromagnetic signals and exacerbates the effect. Universal serial bus (USB) devices are particularly susceptible to EM radiation. The solution proposed was to construct a Faraday cage, a structure designed specifically to counteract the problem of electromagnetic interference and house the reactor inside it.

From [3 and 4] sample calculations were performed to quantify the amount of x-rays produced in a corona discharge-type reactor. 65 kV pulses were applied to a 20 kV DC bias (total 85 kV), the pulse duration was 110 ns and the pulse repetition frequency was 10 Hz.

Over the course of one pulse the x-ray energies peaked at 42 keV. Time resolved intensified CCD images over the duration of a pulse allowed the total number of streamers along the length of the wire to be calculated. Across the 0.9 m wire used in the literature the number of streamers observed was 1260. It was found that over the course of 5795 streamer corona discharges X-rays were observed on only 17 occasions. Therefore, on average, 3.70 X-ray producing streamers are observed.

The total X-ray energy generated per pulse is then the product of the maximum X-ray energy and the probability of an x-ray streamer to be formed. This is equal to 156 keV or 2.49×10^{-14} J.

The maximum dosage of x-rays would be applied to the body with the lowest mass. The mass of the lightest member of the research team was taken as a basis (50 kg).

$$\frac{2.49x10^{-14}}{50} = 4.97x10^{-16} J/kg$$

Taking the pulse frequency into account a more realistic calculation of the X-ray dosage is found. i.e., an 85 kV pulse 110 ns in duration repeating every 0.1 seconds.

$$\frac{4.97x10^{-16} J/kg}{0.161\ hr} = 3.09x10^{-15} S_v/hr \ or\ 3.09x10^{-9} \mu S_v/hr$$

This value was then compared to background radiation level and it was found that working in close proximity to the corona reactor would increase exposure to radiation by 1.00×10^{-7} %, an insignificant amount. Therefore, it can be concluded that the corona reactor is safe to operate on a continuous basis.

4. Sample Results and Proof of Concept

During commissioning a number of initial experiments were performed to ensure all equipment was functioning. Data were collected using the set up seen in Fig. 1. An example of the results obtained can be seen in Fig. 2. Conversion of CO_2 ranged from 1.00 to 14.08% under a number of different conditions (voltage 6-40 kV, frequency 600-2000 Hz and CO_2 inlet composition 9-33%). The parameters altered during testing were voltage and frequency of the power supply together with the gas composition.

Early trends identified were that increasing voltage and frequency had a positive effect on CO_2 conversion. Voltage was observed to have a greater effect on conversion.

5. Conclusions

Development of the experimental system has been completed. Pairing of the power supply with the reactor proved problematic at the small scales being investigated in these studios. However, stable corona has been demonstrated.

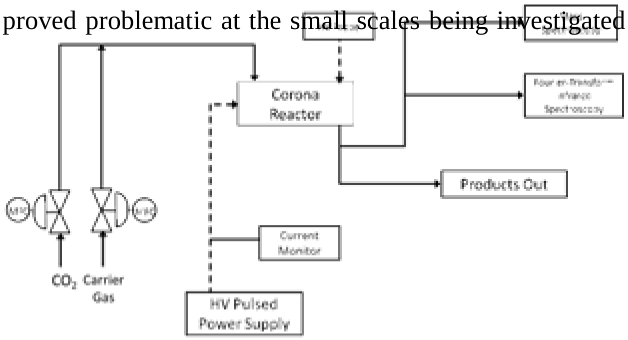

Fig. 1. Schematic of experimental set up.

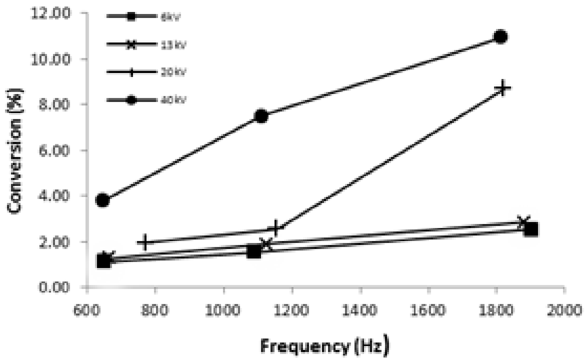

Fig. 2. Conversion of a 33% CO_2 mixture in argon.

Problems with electromagnetic interference were identified during commissioning and a Faraday cage to counteract these problems was designed and constructed. Additionally, the potential for X-rays to form in the corona reactor was identified and sample calculations undertaken to determine that no extra precautions were needed.

Successful ignition of a corona plasma within the reactor was observed in both pure CO_2 and CO_2/Argon mixtures with the setup. During plasma operation CO_2 was successfully reduced to CO with comparable conversions to early DBD technologies.

6. Acknowledgements

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7. References

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