Electronic Supplementary Material (ESI) for RSC Advances

Nonthermal plasma assisted co-processing of CH_4 and N_2O for methanol production

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In a DBD reactor, the discharge was generated in a cylindrical quartz tube with an inner diameter of 20 mm. A copper wire rolled on the outer surface of the quartz tube acts as the outer electrode, whereas a stainless steel rod was used as the inner electrode (Fig. S1). The discharge length was 15 cm and discharge gap was fixed at 3.5 mm. The inner electrode was connected to AC high voltage source (Yaskawa varispeed F7 AC inverter, AC voltage 0-40 kV and frequency 50-1000 Hz variable), whereas the outer electrode was grounded. The discharge was ignited by applying the AC high voltage in the range 12–20 kV (peak–peak) at 50 Hz. The flow rate of gas was controlled with mass flow controllers (GFC-17, Aalborg-USA).

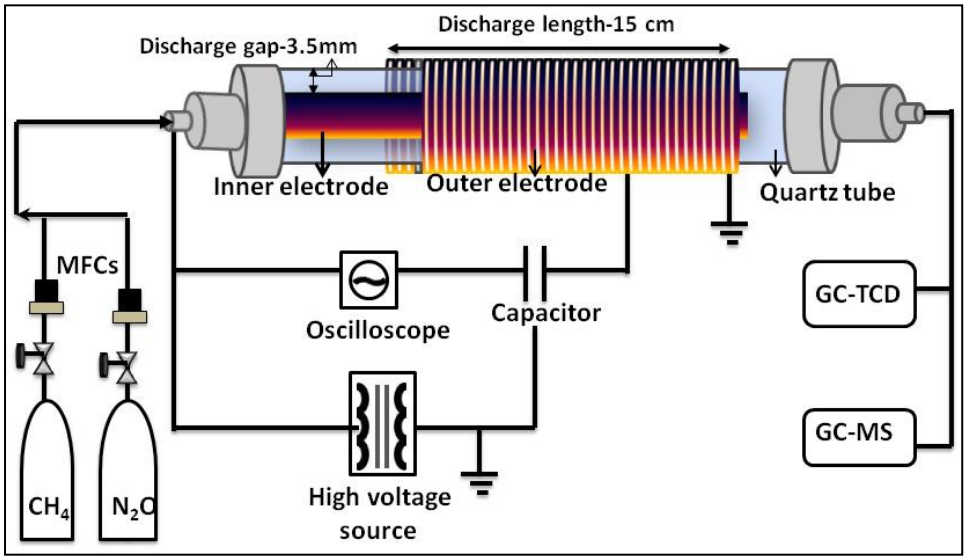

Figure S1. Schematic representation of NTP-DBD reactor

The electrical power applied to the discharge in the DBD reactor was measured by using the V-Q Lissajous diagram, where the charge Q (i.e., time integrated current) was recorded by measuring the voltage across the capacitor (C = 1.5 μF) connected in series to the ground electrode. Applied voltage was measured with a 1000:1 high voltage probe (Agilent 34136A) and the V–Q wave forms were monitored by a digital oscilloscope (Tektronix TDS 2014 B). The area of Lissajous figure (Figure S2) characterizes the energy dissipated during the one period of voltage, from which power was calculated by multiplying with frequency. Specific input energy (SIE) of the discharge was calculated by following relation,

$$\text{SIE (J/L)} = \frac{Power(W)}{Gasflowrate(L/Sec)} \text{ x } 100$$

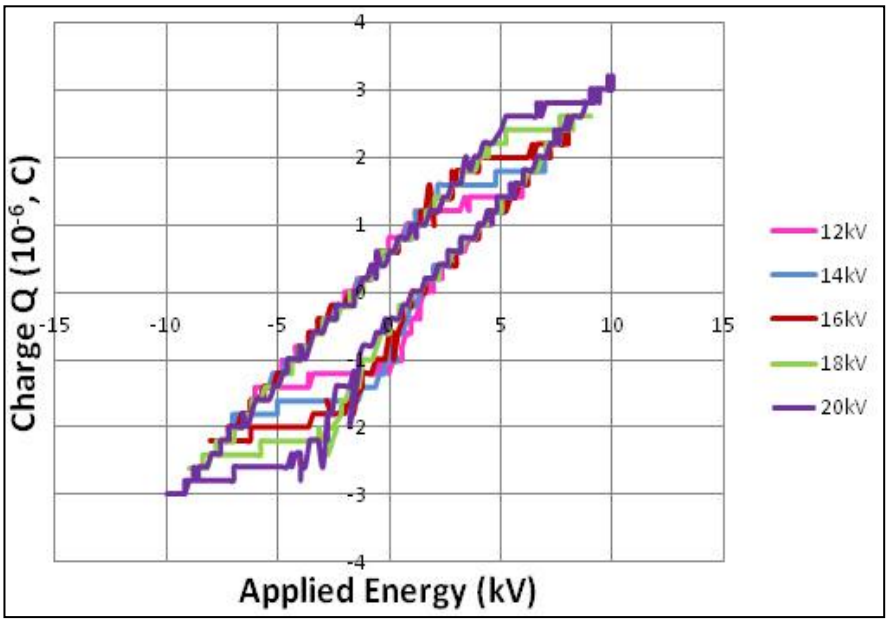

Figure S2. V-Q Lissajous diagrams of 12-22 kV taken at 3.5 mm discharge gap and 50 Hz frequency for feed ratio of CH_4/N_2O-5:1

The gases used in the present study are high pure CH_4 (10% CH_4 diluted in argon) and N_2O (10% N_2O diluted in argon). CH_4 and N_2O were introduced into the discharge zone through a Teflon tube, whose concentrations along with the products (Methanol formaldehyde and hydrogen) were measured with a gas chromatograph (Varian 450 GC) equipped with two TCD detectors, whereas an infrared CO_x analyzer (AIC, India) was used to monitor the CO and CO_2 formed in the reaction. Concentration of hydrogen was confirmed with a hydrogen gas analyzer (Siemens, calomat 6E), whereas, other hydrocarbon products were identified by using GC-MS (Thermofisher). The selectivity of methanol, formaldehyde, hydrogen CO_2, CO and total carbon selectivity was defined as follows,

$$\text{Conversion of CH4}\,(\%) = \frac{[CH4]out}{[CH4]in} \times 100$$

$$\text{Conversion of N2O}\,(\%) = \frac{[N2O]out}{[N2O]in} \times 100$$

$$\text{Selectivity of CH3OH}\,(\%) = \frac{[CH3OH]out}{[CH4]in} \times 100$$

$$\text{Selectivity of HCHO}\,(\%) = \frac{[HCHO]out}{[CH4]in} \times 100$$

$$\text{Selectivity of CO}\,(\%) = \frac{[CO]out}{[CH4]in} \times 100$$

$$\text{Selectivity of CO2}\,(\%) = \frac{[CO2]out}{[CH4]in} \times 100$$

$$\text{Selectivity of H2}\,(\%) = \frac{[H2]out}{2[CH4]in} \times 100$$

$$\text{Total carbon selectivity}\,(\%) = \frac{[CH3OH]out + [HCHO]out + [CO]out + [CO2]out}{[CH4]in} \times 100$$