Mass Spectrometry Study of Destruction of Methane by a Radio Frequency Discharge

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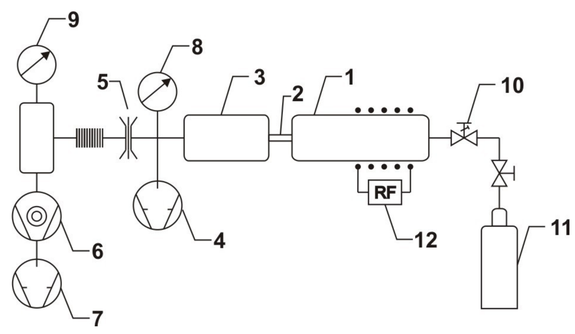
ABSTRACT

In order to understand the kinetics of a-C:H formation in laboratory radio frequency (RF) plasma reactors for scavenger experiments, destruction of methane was studied in a low pressure electrodeless RF discharge. The plasma was ignited in pure methane at flow rates ranging from 25 sccm to 140 sccm by means of a RF generator capable of delivering up to 1000 W output power. The generator was coupled to the experimental system by means of a 6 turn, 4 cm diameter coil. Depending on the output power and gas flow rate, the system was operating either in E or H mode. The composition of the discharge was analysed by means of mass spectrometry. It was found that the destruction of methane depended strongly on the discharge power, gas flow rate and operating mode. Radicals created by the discharge were found to recombine and form bigger molecules, such as ethylene and acetylene.

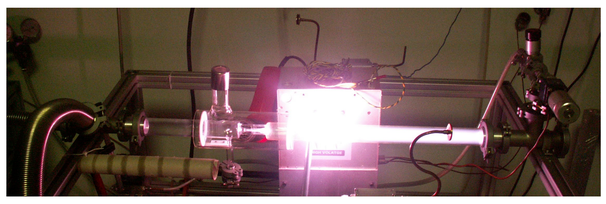
Experimental system



The experimental system consisted of a quartz tube with the inner diameter of 36 mm. Methane of commercially avaliable purity (11) was continuously leaked in through a mass flow meter (10) and the system was pumped with a two stange rotary pump (4) with the flowrate of 80 m³/h. The pressure was measured with a Baratron gauge (8).

Plasma was ignited and sustained by means of a 13.56 MHz generator (12), capable of delivering up to 1000 W output power. It was inductively coupled to the discharge chamber (1) by means of a matching network and a 6-turn coil. In order to prevent propagation of charged particles into the afterglow chamber (3), the two chambers were separated by a 40 mm long narrow tube with the inner diameter of 3 mm (2). The afterglow chamber was terminated by a shutter (5). Depending on the nominal power and methane flowrate, the plasma reactor was operating either in E or H mode.

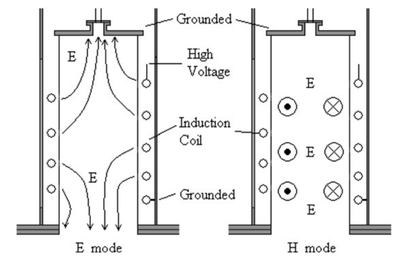
A residual gas analyzer (9) was used to monitor the partial pressure of reaction products of the plasma. Due to the relatively high working pressures, it was differentially pumped by a turbomolecular pump with the pumping speed of 250 l/s (6), backed by a two stage rotary pump with the pumping speed of 8m³/h (7).



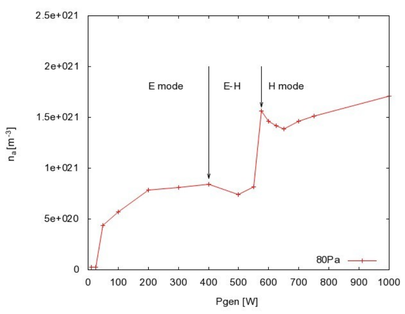
A photograph of the experimental system. Note that the plasma is elongated all along the discharge chamber which indicates that the system is operating in E-mode.

E – H mode transition

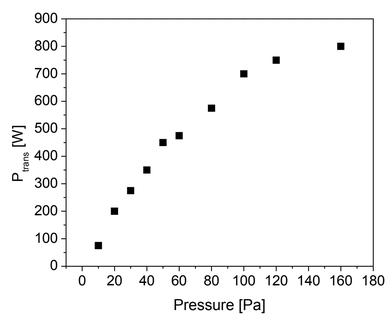
In inductively coupled RF plasmas, there are two modes of operation, the E-mode and the H-mode. In the E-mode, the plasma is sustained predominantly by an electrostatic electric field whereas in the H-mode it is sustained by the induced electic field.



In our reactor, the transition from E-mode to H-mode occurs when increasing the reactor power. In the transition, the electron density increases drastically, while the electron temperature drops slightly. The transition between the E- and H-mode is also very pronounced in the density of atomic oxygen produced in an O_2 discharge. Typically, as the working pressure increases, the E-H transition occurs at higher powers.



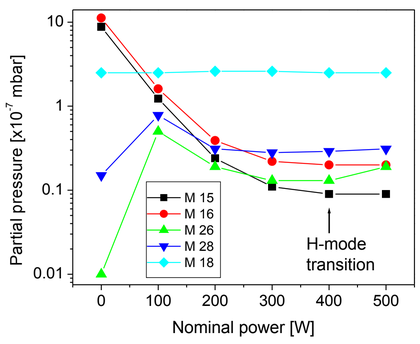
Atomic oxygen density vs generator power, recorded at the source gas pressure of 80 Pa. The transition to H-mode occurs at 600 W.



Generator power where the transition to H-mode occurs versus source gas pressure, for oxygen plasma.

Results

25 sccm

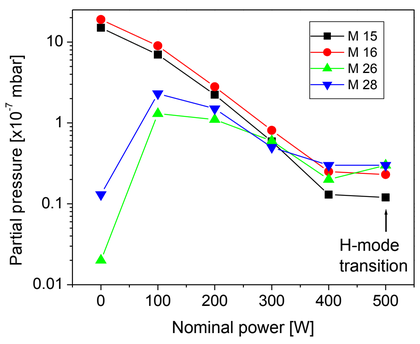


Partial pressure of methane (masses 15 and 16) decreases rapidly and reaches saturation value before the E-H transition at 400 W, which suggests total destruction of CH_4.

Ethylene (mass 28) and acetylene (mass 26) appear as reaction products. The peak at 100 W suggests that at higher powers, the degree of dissociation of C_2H_4 and C_2H_2 increases.

The E-H transition has little effect on stable reaction products.

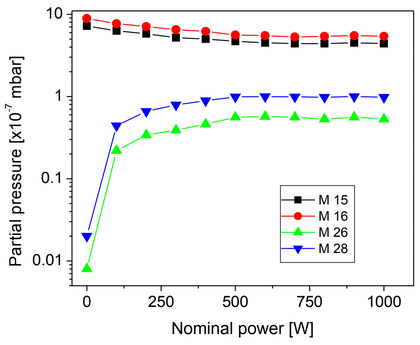
40 sccm



Total destruction of methane shifts to 400 W. This indicates that, at a higher flow rate of CH_4, more power has to be invested into the discharge before the precursor is completely depleted.

The peaks of C_2H_4 and C_2H_2 also broaden towards higher powers which suggests that in this case, due to a higher density of precursors, the rate of production competes with the rate of dissociation more easily.

80 sccm

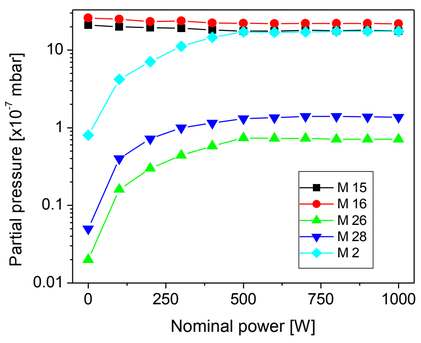


At this flow rate, the complete destruction of methane is not achieved. The CH_4 partial pressure decreases only by approximately a factor of 2, even at generator powers up to 1000 W.

Similarly, the partial pressures of ethylene and acetylene have no apparent peak, but exhibit asymptotic growth which reaches the saturation value at the same power as the CH_4 partial pressure.

At this flow rate, the E-H transition is not observed.

140 sccm



The situation doesn't change much when we further increase the flow rate. It is also interesting to observe the partial pressure of H_2 (mass 2), which is one of the stable reaction products of the discharge, too. It exhibits the same behaviour as C_2H_4 and C_2H_2.

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