Hydrogen production from ethanol decomposition by a surface wave discharge at atmospheric pressure

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Surface wave plasma at atmopsheric pressure has been used to produce the decomposition of ethanol molecules introduced into it, in order to obtain molecular hydrogen at the plasma exit. The main gaseous products obtained were H_2, CO and C_2H_2 whereas H_2O, O_2 and CO_2 were not formed during the ethanol reforming process. The concentration of the main products increased when the Ar flow used as plasma gas decreased.

Here, we present a specific study on the ethanol decomposition by a surface wave plasma at atmospheric pressure for obtaining molecular hydrogen at the discharge exit. The plasma was a gas mixture of argon and ethanol. The influence of Ar flow (in the mixture) on H_2 production, keeping constant the amount of alcohol (g/h), was analyzed. The success of both the dissociation and excitation processes of molecules by the plasma particles depends on the electron density and the energy available in the discharge which is mainly provided in the form of the kinetic energy of plasma particles (electrons and heavy species). Optical Emission Spectroscopy (OES) techniques were used to determine the plasma parameters, gas temperature and electron density, which are considered the key parameters to induce the internal chemistry reactions in non-equilibrium plasmas.

Microwave power (200W) in a continuous mode was supplied for plasma creation by means of a 2.45 GHz SAIREM microwave generator (GMP 03k/SM). A surfatron [1] was used as the energy coupler device. Argon gas (purity ≥ 99.999%) flows of 0.50, 0.75, 1.00 and 1.25 were used for generation of plasmas created in a quartz tube of 1.5 mm and 4 mm inner and outer diameter, respectively. Straight after plasma ignition with the Ar gas, an ethanol flow of 0.22 g/h was added to the plasma by using a gas phase liquid delivery system (CEM, system Bronkhorst). The ethanol vapor/argon mixture was led to the plasma through a steel tube heated at 110ºC to maintain its temperature over the ethanol boiling point so as to avoid alcohol condensation which could produce the plasma extinction. Due to the plasma high temperature, the quartz tube was externally refrigerated by an air flow from high-pressure air tank at room temperature.

Two types of analysis were carried out. On one hand, the plasma capability of inducing the decomposition of ethanol molecules was analyzed through the radiation emitted by the plasma. This radiation was collected perpendicularly to the discharge tube at 2 cm of the end of the plasma column (in all cases) by a PCS 1000 optical fiber with a silicon core of 1000±40 μm diameter and guided to the entry slit of a Czerny-Turner monochromator (Jobin-Yvon Horiba) with 650 cm of focal distance and gratings of 1200 and 2400 lines/mm. A Hamamatsu R928P photomultiplier (PMT) and a Symphony CCD were used as radiation detectors. Spectra recorded permitted us to gain information about the different species existing in the plasma and to estimate the gas temperature (CCD camera) and the electron density (PMT) in the discharge. On the other hand, a quadrupole mass spectrometer (PT M63 112, mod. Omnistar) was placed on line to the discharge tube to detect the produced molecular hydrogen.

The variation of the gas temperature with the Ar flow in the argon-ethanol mixture is shown in Figure 1a. As it can be observed, the gas temperature presents a tendency to decrease when the Ar flow increases from 0.5 to 1.25 slm. In Figure 1b, the electron density values obtained from the Stark broadening of H_α line at different electron temperatures from Gigosos and Cardeñoso computational model [2] and the calibration method [3] are depicted. One observes, in all cases, a tendency to decrease when the Ar flow increases. likewise, the values obtained from the calibration method and the $\Delta\lambda_S(H_\alpha)$ for an electron temperature of 5000 K are closest. This electron temperature is characteristic of plasma columns generated with Ar gas by a surface wave at atmospheric pressure [4].

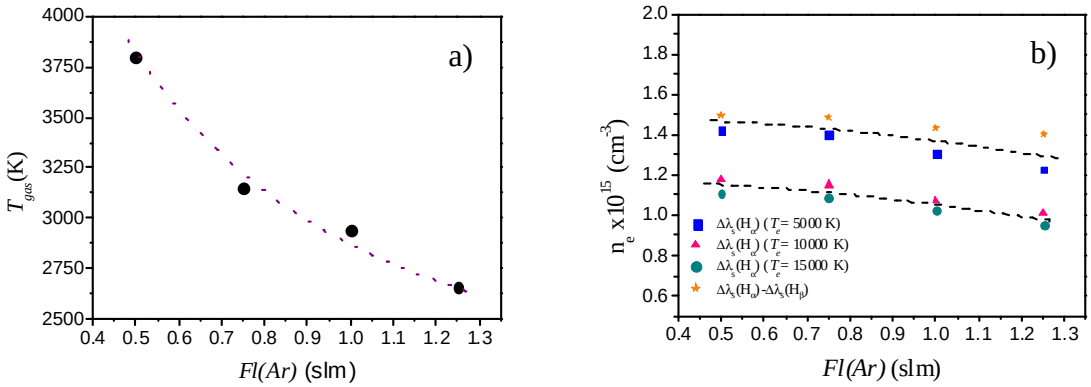

Fig.1: a) variation of the gas temperature with the Ar flow in the argon-ethanol mixture and b) electron density value calculated from Stark broadening of H_α line for the different Ar flows.

Figure 2a shows the mass fragment peaks between m/e = 1 and 60 obtained for the reactive mixture Ar-ethanol with the plasma switch on and off.. Ethanol was totally transformed in the plasma reactor giving rise to conversion close to 100 %. The main gaseous products obtained were H_2, CO and C_2H_2 (H_2>CO>C_2H_2). Besides, trace amounts of C_4H_2 were also detected, while other fragments peaks related to saturated hydrocarbons or higher hydrocarbons were not produced. On the other hand, a small amount of carbon deposited and belched from the quartz tube was also collected. Figure 2b shows the ion current intensities measured in the mass spectrum, at the plasma exit, for ethanol, H_2, CO, C_2H_2, CO_2, H_2O and O_2 as a function of the time for different flows of Ar gas as plasma carrier gas. On one hand, one observes that the concentration of H_2, CO and C_2H_2 increases when the argon flow decreases. On the other hand, note that the measured intensity for H_2O, O_2 and CO_2 do not change with the gas flow rate since they are not formed during the ethanol reforming process.

These results show the capability of surface wave plasma generated at amopsheric pressure to decompose the ethanol molecules and obtaining molecular hydrogen at the plasma exit.

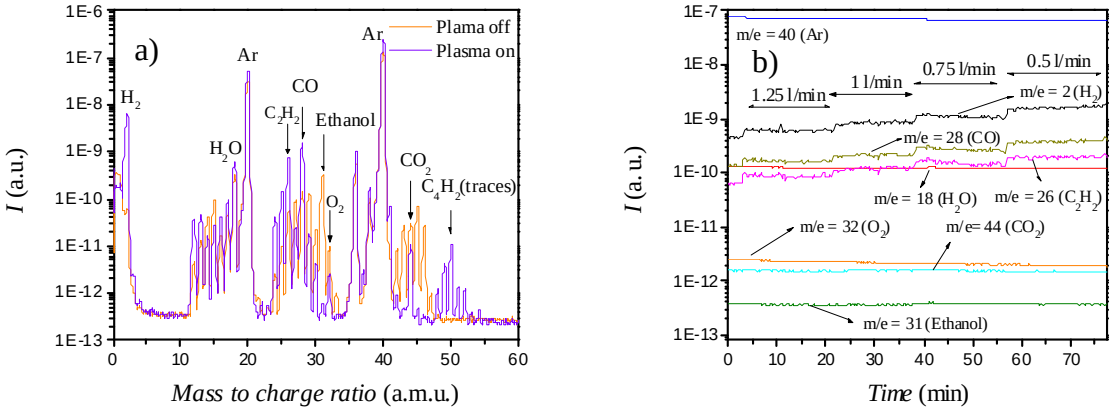

Fig.2: a) mass spectrum of the argon-ethanol mixture with the plasma on and off, b) change of the mass spectrum versus time for the four Ar flows studied.

References

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