An alternative source to produce hydrogen from alcohols by using an argon surface wave sustained discharge at atmospheric pressure

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In view of the need to find alternative sources to produce energy that could be considered to be profitable and that do not emit polluting gases, this study presents a procedure to obtain hydrogen. For this purpose, alcohols (methanol and ethanol) have been introduced into an argon surface wave sustained discharge (SWD) at atmospheric pressure. In this way, particles within the plasma, break the bonds of the alcoholic molecules, liberating the hydrogen contained in them. The SWDs have some characteristics that offer many advantages for this purpose; the microwave energy is coupled to the discharge using an excitation device placed outside the plasma tube, and in this way long plasma columns are obtained. In addition, very low microwave power is required for SWD generation, obtaining values of the electron density and temperature similar to those provided by other types of discharges.

1. Introduction

Due to the problems related to traditional energy sources, the decrease in the natural reserves of fossil combustibles or the contamination that current processes for energy generation provoke (CO₂ emission or the hazards of nuclear power sources), it is necessary to look for alternative ones, like the use of *hydrogen* which is becoming more and more prevalent. An argon surface wave sustained discharge (SWD) at atmospheric pressure has been used in order to obtain hydrogen from alcohols (methanol and ethanol), since these discharges have been considered to be an effective source of reactive species used in quite a lot of chemical processes, some of them being hardly carried out by the standard chemistry [1-2].

2. Experimental device

The discharge was generated inside 1.5 and 4 mm inner and outer diameters, respectively, the quartz tube opened at one end, using a total gas flow of 0.5 slm with a purity of 99.999% (impurities were mainly water and nitrogen) and a microwave power of 200 W to generate the plasma. This power was supplied by a surface *surfatron* wave launcher [3] (Figure 1).

A method called *bubbling* (Figure 2) was employed for the introduction of the alcohols into the plasma. The total gas flow was divided into two parts; one of them used as carrier gas to drag the volatile phase of the alcohol to be introduced into the plasma. This argon flow was then united to the rest of the argon flow before introducing the total flow into the quartz tube which contains the discharge. Through the contact between argon gas

and liquid alcohol, the transport towards the discharge is favoured.

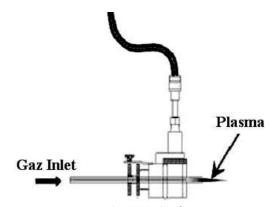


Figure 1. Surfatron.

The possibility of the sample excitation processes depends on the energy available in the discharge, mainly in the form of the kinetic energy of the electrons and the heavy particles, such as argon atoms and ions, which transmit their energy to the particles of the samples by collisions. Thus, the alcoholic molecules, inside the plasma, are easily broken and excited through collisions with the plasma particles.

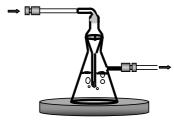


Figure 2. Method of bubbling.

3. Results

Light emitted from the plasma was guided through an optical fibre to the entrance slit of a 1 m Jobin-Yvon Horiba monochromator. A computer-controlled CCD camera was used to register the spectra. In this way, the H_{α} Hydrogen Balmer Series line was analyzed by Atomic Emission Spectroscopy (AES), resulting in an increase in their intensities (Figure 3).

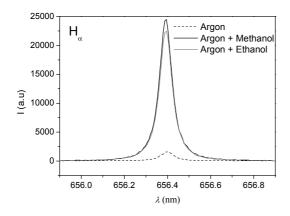


Figure 3. Balmer Series H_{α} lines registered from the plasma discharge.

A preliminary study has also been carried out at the exit of the plasma employing a mass spectrometer, and an increase in the production of molecular hydrogen has been observed when the alcohols have been introduced into the argon plasma.

Not only the Hydrogen Balmer Series lines have been monitored; it is also possible to detect rotational bands corresponding to the C₂, CN, NH, and CH species. This demonstrates that the bonds between carbon and hydrogen atoms in the alcohols are being broken. In addition it has been verified that all argon lines, except for 495.73 nm, decreased after sample introduction. The average electron energies on this kind of discharges are 2-4 eV and the ones of different molecular bonds are C-C: 3.6 eV, C-O: 3.7 eV, C-H: 4.3 eV and O-H: 4.7 eV. Observing the energies, we can conclude that molecular bonds, except for the O-H, are easily broken by the plasma particles (electrons) permitting the excitation of the different molecular fragments.

Among the bands detected, the one corresponding to the C_2 specie (Figure 4) can be considered very important due to the application to produce carbon nanotubes (molecular-scale tubes of graphitic carbon that show outstanding properties).

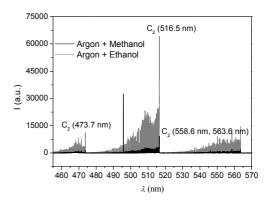


Figure 4. Bands corresponding to the C_2 specie

So, we can summarize that the global reaction occurring in the plasma is given for:

$$C_n H_{2n+2}O \rightarrow nC + nH_2 + H_2O$$
 (1)

This study has been carried out at reduced pressures by Yaguas-Gil *et al.* [4]. These authors have observed that at reduced pressure the CO emission is emitted unlike it happens at atmospheric pressure, where this emission has not been observed (Figure 5). This is the reason why we think that it could be an non-contaminating alternative source.

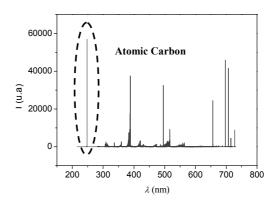


Figure 5. Ar + Methanol emission spectrum at atmospheric pressure.

4. References.

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