Hydrogen production from alcohols using a surface wave discharge (SWD) at atmospheric pressure

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Abstract

In this work the results in the H_2 production from alcohols by microwave plasma at atmospheric pressure are presented. The importance of this study is based in the need to find other energy sources as an alternative to the oil use.

The results show that the process presents a great simplicity, opposite to other complicated systems of generation of hydrogen and a low energetic cost compared with the other alternative energies.

Keywords: Hydrogen production; Microwave plasmas; Atmospheric pressure

1. Introduction

In view of the problems related to the traditional energy sources it is necessary to look for alternative sources of energy among which the usage of *hydrogen* is gaining more and more importance. Due to the relative shortage of H_2 free in our atmosphere (0.01 % in volume of the pure air), this compound must be obtained from the break of molecules that contain it in their composition, in general from great part of the hydrocarbons. However, it is necessary to use an effective source of energy to break the links of the above mentioned molecules, in order to liberate the present hydrogen in the same ones (reformed). Nevertheless, the systems used to produce H_2 nowadays, such as methane reforming, are expensive and yield CO_2 (contaminating gas) emission. Therefore, it is necessary to find an alternative H_2 production method, energetically profitable and without the emission of this contaminating gas.

The plasma characteristics, such as high temperature, energy density and degree of ionization, fast response time and minimal cost, have led several researchers to use plasma discharges as a reactive media to H_2 production [1-2]. The plasma can effectively provide the energy required for endothermic reactions of reforming. The main objective of our study is to achieve H_2 production from alcohols, due to their high H_2 composition, in a clean way by using an argon plasma created and maintained by a microwave surface-wave (SWD) at atmospheric pressure.

2. Experimental procedure and Results

The argon discharge was created in a quartz tube with one of its ends open, obtaining a plasma column at atmospheric pressure. The dimensions of the inner and outer diameters of the discharge tube were 1.5 and 4 mm, respectively. This diameter has been chosen because it is known that a big diameter originates more than a plasma filament in the tube cross section, providing a colder plasma-free zone where the fragmented hydrocarbon molecules decrease. The plasma gas was argon with a purity of about 99.99% at a flow of 0.5 slm controlled by a HI-TEC system (model IB 31). The microwave power for the creation and maintenance of the plasma was 120 W and provided by a SAIREM generator (GMP 20 KE/D) in a continuous mode at a frequency of 2.45 GHz. This power was supplied to the plasma by a *surfaguide* [3] as an exciter device.

The result analysis is carried out by studying the radiation emitted by the plasma, which is collected in the shape of spectrums by an optical fiber. The fiber was conducted up to the entrance slit of a Jobin-Yvon Horiba monochromator. By using this one, the radiation was decomposed into different optical wave lengths (spectral lines), which corresponded to the different elements present in the discharge. Figure 1 shows the schematic diagram in which appear the plasma source and the optical configuration to register the spectra.

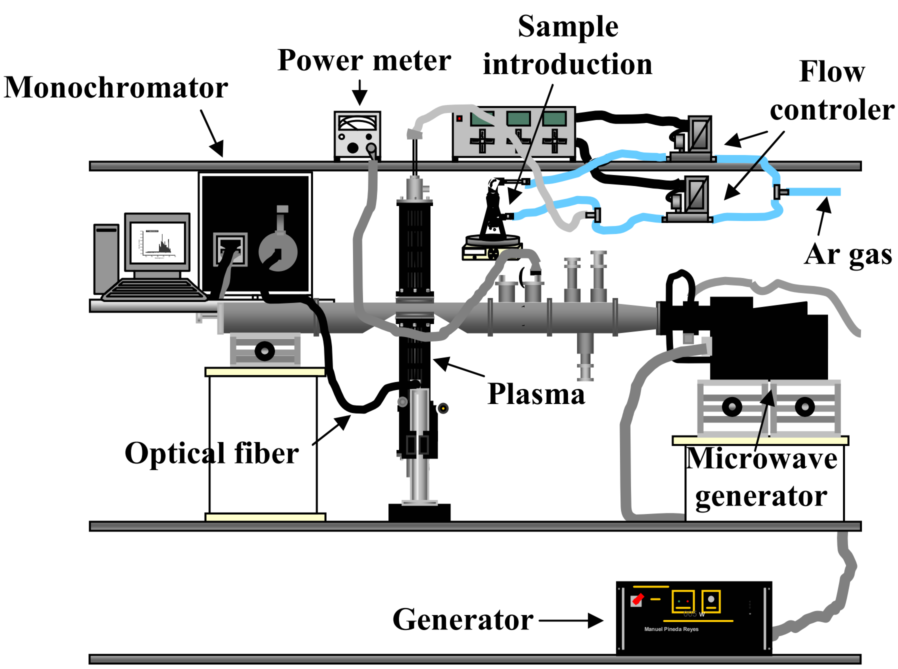

Figure 1. *Diagram of the experimental arrangement*

The hydrocarbon samples (methanol, ethanol, propanol and butanol) were put into the plasma gas (argon) flow through two systems, novel at the moment of introducing liquid (volatile) samples in this type of discharge, and that are characterized because they do not involve in the process an energetic additional significant consumption. In the first system, named *sweeping*, the samples are warmed softly and once evaporated they are dragged by the argon up to the plasma. In the second system, the gas *bubbling*, that takes place at environment temperature, the gas argon was bubbled in the sample of alcohol, dragging molecules of sample towards the gas flow. This mixture, already formed by the gas flow and the sample, was mixed by the rest of the flow of the argon and later they were injected together into the discharge, increasing this way the reaction time of the species before being introduced in the plasma. When the hydrocarbons were inside the plasma, their molecules interacted with the plasma particles which transferred efficiently, by means of collisions, the necessary energy to break the links of the introduced molecules. The energy of electrons in this discharge type (microwave discharge) is of about 4 eV, which is not high enough to break these links, whereas that of the levels metastables and resonant of the argon it is of about 11 eV, so these particles being as a reserve of energy to carry out these reactions [4].

The abundance of the produced hydrogen was related with the area of the spectral lines of the hydrogen registered, such as H_α (656.28 nm) and H_β (486.13 nm). In Ar pure plasma the hydrogen was present as impurity. In Figure 2, we can observe that the intensity of the H_α lines increased significantly when the different samples of alcohols are introduced. This fact reflects that the molecules of hydrocarbons are breaking inside the plasma, liberating the hydrogen that was forming a part of their composition.

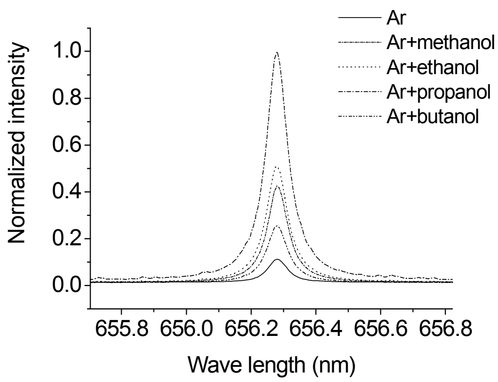

Figure 2. *Intensity of the line H_α registered from the plasma with different samples.*

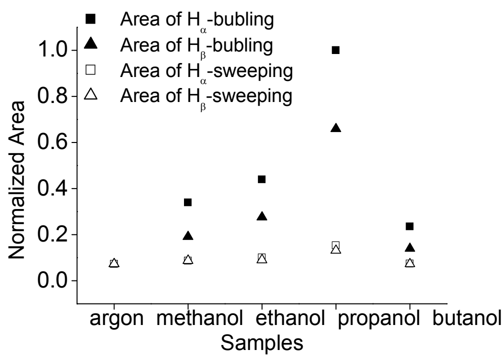

Figure 3. *Normalized intensity of the H_α and H_β lines registered.*

The behavior of the intensity of both lines used (H_α and H_β) in this study with the different samples and with both introduction systems are shown in Figure 3. In this figure the area of the hydrogen lines have been normalized respect to the higher area value obtained for these lines, which corresponded to the H_α line in the case of sample bubbling. One observes that H_2 production depends on the introduced sample, being higher with the propanol. Therefore, H_2 production is greater by using the bubbling method than the sweeping system. Therefore, we can avoid the previous heating required by the second system, which simplifies the sample introduction and reduces the energetic cost.

Finally to point out that only bands corresponding to NH, CN, CH, C_2 and OH species were observed. In any case CO_2 and CO bands were found.

3. Conclusions

There has been verified that it is possible to produce hydrogen inside a discharge of argon generated by microwave power at atmospheric pressure, introducing samples of alcohols, which contain this hydrogen in their composition. The process presents a great simplicity, opposite to other complicated systems of generation of hydrogen and a low energetic cost compared with the other alternative energies. Also, the process is clean, since there has not been detected emission of CO_2, because the plasma is generated inside a quartz tube and it is not in direct contact with the oxygen of the air. In addition, alcohols such as ethanol can be derived from vegetables (cereals or sugar cane). This preliminary work has to been completed with more exhaustive studies in order to find the best conditions of microwave power and gas flow for a maximum hydrogen production consuming the minimal energy.

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