Microwave plasma torches applied for hydrogen production

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Abstract

The operation of microwave plasma torches in Ar-methanol and Ar-methanol-water mixtures at atmospheric pressure for molecular hydrogen production is the purpose of this study. Mass and optical spectroscopy have been used to detect the molecular hydrogen production and to analyze the plasma emission, respectively.

Introduction

In the last decade, hydrogen has attracted great attention as an alternative energy source. Hydrogen gas can be produced from substances containing hydrogen atoms such as hydrocarbons [1]. High-density plasma torches are known to provide suitable conditions to dissociate molecules in the abatement systems and burn out chemical and biological warfare agents as well as to atomize materials. One of the main advantages of microwave plasma torches at atmospheric pressure is that they make it possible to inject large power densities into the plasma and thus achieve high densities of active species of interest. An essential advantage of microwave plasmas over other types of discharges, such as gliding arcs, corona and dielectric barrier discharges, is the absence of electrodes [1-3]. In this way, problems such as electrode erosion, discharge pollution, and additional energy consumption for cooling are avoided.

In this study, we test the ability of a microwave plasma torch to decompose methanol and produce hydrogen. The main aim is to find the optimum conditions for this process i.e. the appropriate gas flow, power and reforming substance (methanol with or without water).

Experimental setup

A microwave plasma torch operating at 2.45 GHz and atmospheric pressure has been used. To this end, a plasma in a gas mixture of argon and methanol, with or without water,

has been created using a surfaguide launcher and a microwave generator delivering a power in the range 0.2-2.0 kW. The discharge takes place in a quartz tube (1.5 cm inner and 1.8 cm outer diameters, respectively) open at one end. The total gas flows used in the experiments range from 500 to 4000 sccm. The argon gas used has a purity of 99.999%. Pure methanol is introduced into the discharge by bubbling of Ar through the alcohol at room temperature as shown in Fig.1.

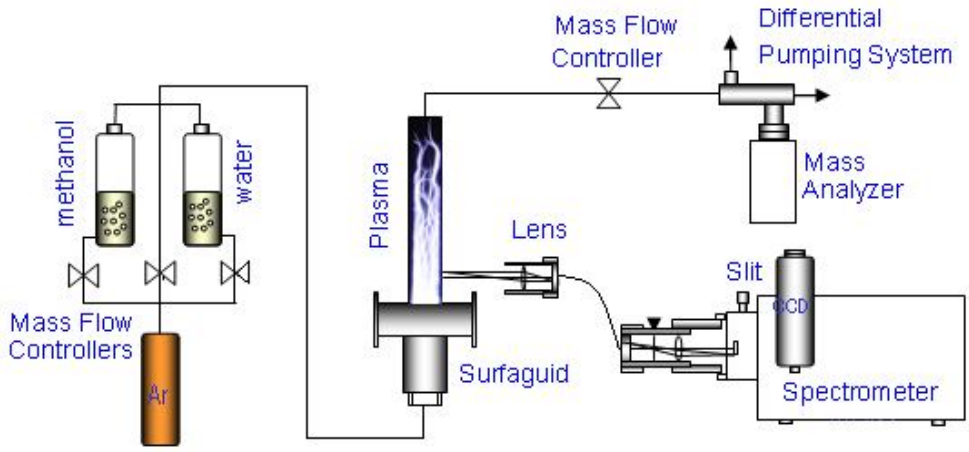

Fig. 1. Experimental setup.

Results and discussion

The time evolution of the H_2 partial pressure detected by the mass spectrometer is presented in Figure 2. The jump of the signal when the discharge is turned on is a clear evidence of molecular hydrogen production from methanol decomposition by the plasma. Besides H_2 and CO, which are the main products of the methanol dissociation process ($CH_3OH \rightarrow 2H_2 + CO$), some H_2O, O_2 and N_2 are detected as well. The influence of the fraction of Ar "bubbling" flow through liquid methanol on the H_2 and CO partial pressures is shown in Fig. 3. There is an increase in both partial pressures with the Ar fraction. The dependence of the produced H_2 flow *vs.* the introduced methanol (Fig. 4) is linear. The obtained ratio for total flows in the range 500 – 2000 sccm corresponds to the maximum theoretical value: i.e., for every introduced methanol molecule two H_2 molecules are formed. Nearly 100 % efficiency of methanol plasma reforming is achieved. The input microwave power is another important parameter in the plasma decomposition process.

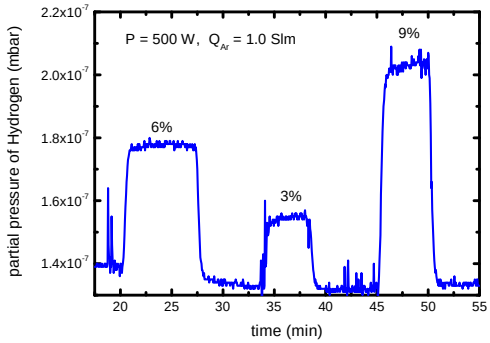

Fig. 2. Time evolution of the H_2 partial pressure as detected by the mass spectrometer.

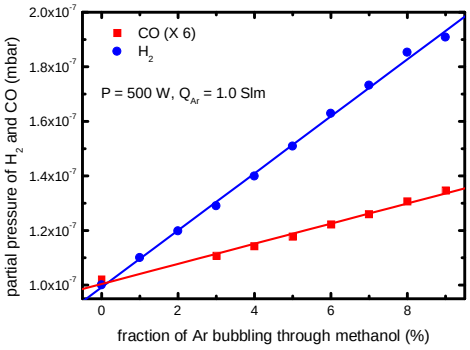

Fig. 3. Dependence of the H_2 and CO partial pressures on the fraction of Ar bubbling through methanol.

The microwave power absorbed per unit volume is linearly correlated with the electron number density. Correspondingly, the results show that the methanol conversion and hydrogen yield increase with the input power (Fig. 5). This can be explained by the fact that more active species are generated due to the higher microwave power. Some saturation in the hydrogen production is however observed for microwave powers greater than 400 W, which for the present conditions corresponds to a power density of about 10 W/cm^3. This suggests that further increase in power density induces a complex chemistry involving dissociation of hydrogen molecules followed by association processes of highly reactive H atoms with other radicals to form hydrogen containing species. This is confirmed by the optical spectra obtained.

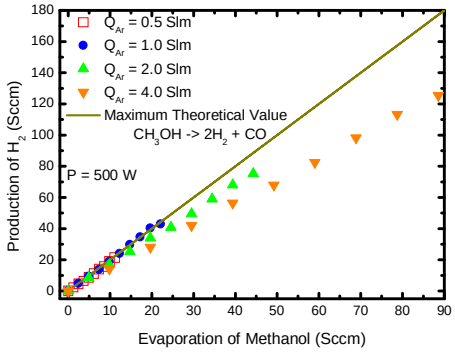

Fig. 4. Hydrogen flow *vs.* pure methanol flux.

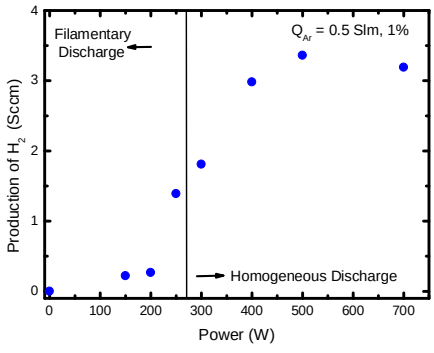

Fig. 5. H_2 flow dependence on the microwave power.

The effect of a small quantity of water vapor on the hydrogen and carbon oxide/dioxide yield has also been investigated. The water in the tank has been heated to about 70°C in order to increase the efficiency of H_2O introduction by Ar bubbling (see Fig. 1). The presence of water contributes to the increase of H_2 and CO_2 (see Fig. 6) by the steam reforming of methanol: $CH_3OH + H_2O \rightarrow 3H_2 + CO_2$. Besides, the methanol decomposition is followed by the water-gas shift reaction $CO + H_2O \rightarrow H_2 + CO_2$, which further increases the yields of H_2 and CO_2. Therefore, if enough water was introduced, all of the carbon oxide would be transformed into carbon dioxide.

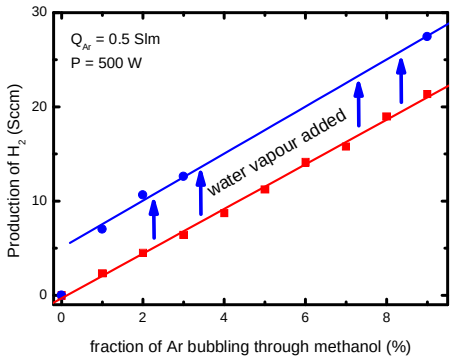

Fig. 6. Produced H_2 *vs.* Ar "bubbling" flow through methanol.

Conclusion

This work clearly shows that the present microwave torch can be very efficient for H_2 production from methanol. The best results were obtained for low argon flows (up to 1000 sccm). The optimal microwave power is about 400 W. Addition of water increases the H_2 production but high H_2O flows influence the discharge stability.

References

[1] J.D. Holladay, J. Hu, D.L. King, Y. Wang, Cat. Today, 139, (2009), 244–260

[2] B. Sarmiento, J. Javier Brey , I.G. Viera , A.R. Gonzalez-Elipe, J. Cotrino, V.J. Rico, , J. Power Sources 169, (2007), 140–143

[3] M. Jimenez, C. Yubero and M.D. Calzada, J. Phys. D: Appl.Phys. 41 (2008)

[4] G. Petitpasa, J.-D. Rollier, A. Darmonb, J. Gonzalez-Aguilar, R. Metkemeijer, L. Fulcheri Int. J. Hydrogen Energy, 32, (2007), 2848–2867