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SCIENCE DIRECT®
Surface & Coatings Technology 200 (2006) 6434 – 6437
SURFACE & COATINGS TECHNOLOGY
www.elsevier.com/locate/surfcoat

Growth of thin hydrocarbon films and hydrogen production in a cusp plasma device

N. Spinicchia [a,*], G. Angella [b], M. De Angeli [a], G. Gervasini [a], E. Signorelli [b]

[a] *Istituto di Fisica del Plasma "P. Caldirola" Assoc. EURATOM-ENEA-CNR, Milano, Italy*
[b] *Istituto per l'Energetica e le Interfasi CNR, Milano, Italy*

Available online 20 December 2005

Abstract

A plasma device with a cusp magnetic field configuration and operating under steady state condition has been built to investigate surface deposition of hydrocarbon films at low temperature and, simultaneously, the cracking of methane in a plasma environment to hydrogen production.

The gas (methane or a methane/argon mixture) has been fed to the plasma source (a cylindrical capacitively coupled rf type). The addition of a static magnetic field has increased the particle confinement time and, consequently, the hydrogen production rate.

Here the preliminary results concerning the growth of thin hydrocarbon films and the dissociation of methane in the cusp plasma are presented. In our case, at the line and point cusp, we have a large flux of energetic particles as well as neutral, ions, thermal radicals that participate in the growth process.

A fraction of the converted methane has been deposited on the substrates, at the line cusp, as amorphous hydrocarbon (a-C: H) film and amorphous carbon (a-C) film. The surface morphology of the film has been observed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) has been used to determine the structure of the film. The results have indicated that the film has a prevalently amorphous structure with a presence of crystalline clusters.

Keywords: Methane conversion; Hydrogen production; Plasma; Cusp; Hydrocarbon film

1. Introduction

Hydrogen is supposed to have an important role in the future worldwide energy vector supply and environmentally safe technologies. In the last years, plasma has become a great alternative for hydrogen production [1]. The addition of a static magnetic field increases the particle confinement time and, consequently, the hydrogen production rate. In this view, a plasma device with a cusp magnetic field configuration has been built to investigate the cracking of methane in a plasma environment.

The cusp magnetic configuration approach is very attractive for the purpose of cracking of methane. It has a very simple geometry and the high magneto-hydrodynamic stability of the plasma allows to obtain high plasma concentrations with a high dissociation of the CH_4 molecules. Besides technique for plasma processing (e.g. radiofrequency ion plugging [2] or hydrogen recovery by permeation membranes [3]) can be easily combined in order to increase the confinement time and allow the recovery of the converted H_2. They are proposed as forthcoming improvements of the device.

Here the preliminary results concerning the dissociation of methane in the cusp plasma, measured by quadrupole mass spectrometers are presented. A large fraction of the converted methane has been deposited along the line cusp vacuum wall as amorphous carbon (a-C) film. The surface morphology of the film has been observed and the results indicate that the film has a prevalently amorphous structure with a presence of crystalline clusters.

2. Experiment layout

The experiments have been carried out on the cusp plasma device, developed at the Institute of Plasma Physics in Milan. The cusp magnetic field configuration is obtained by two

* Corresponding author.
E-mail address: spinicchia@ifp.cnr.it (N. Spinicchia).

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doi:10.1016/j.surfcoat.2005.11.006

identical bundles of coaxial solenoids where each bundle carries the same electric current intensity but with opposite directions. The static magnetic field can reach a maximum intensity of 0.5 T at the point cusp and 0.3 T at the line cusp. The solenoids are water-cooled allowing continual operation of the plasma device. The experimental apparatus with the vacuum system is shown in Fig. 1a (the electromagnets are depicted by dashed lines). Fig. 1b shows the schematic arrangement of the experimental device.

The plasma source (labeled PS in the figure) is located at one point cusp and is fed with gas (a mixture of Ar and CH_4) through a mass flow controller (FC, model MKS M-200) that maintains a discharge pressure between 10^{-1} and 1 Pa under a flow rate spanning from 2 to 40 sccm. The plasma source is a cylindrical rf capacitor composed by an inner tube with 10 mm diameter, 460 mm length, and an outer electrode (ground) represented by the vacuum chamber tube (internal diameter of 60 mm). The plasma source is powered by a commercial 13.56 MHz rf generator with a maximum power of 300 W (Huttinger mod. PFG 300 RF) connected to an automatic impedance matching network.

At the second point cusp, opposite to the plasma source, is situated a permeation membrane holder (PR), which is placed at a proper distance from the cusp centre, to allow the hydrogen-enriched plasma to strike on the membrane itself. In particular, the presence in the plasma of hydrogen in atomic form will increase the hydrogen permeation through the membrane [3]. The holder is arranged to mount a 60 mm diameter membrane and is provided with heaters and a thermocouple to monitor the membrane temperature. The permeation membrane has a selectivity for hydrogen only.

A Langmuir probe (LP), located along the line cusp, provides plasma parameters such as the electron temperature and the plasma density.

The gas composition at the exits of the line cusp chamber and at the second point cusp can be determined by two quadrupole mass spectrometers (QMS1, QMS2) type Balzers QMA 125 adequately screened against the magnetic field [4]. Here

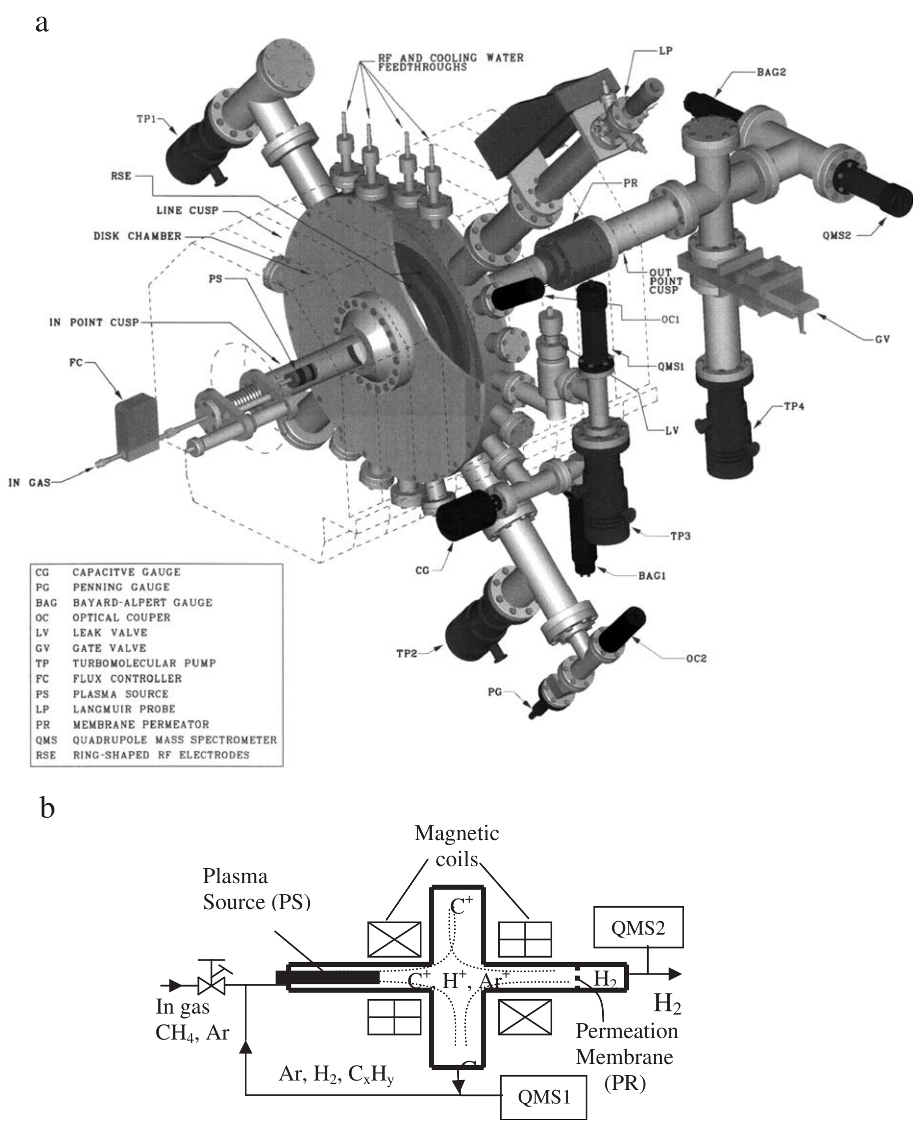

Fig. 1. 3D layout (a) and schematic view (b) of the apparatus.