

Production of hydrogen molecules by ethane and ethene interaction with hot tungsten

I. Čadež, S. Markelj, Z. Rupnik

Jožef Stefan Institute, Association EURATOM-MHEST

Jamova cesta 39, 1000 Ljubljana, Slovenia

Introduction

Use of metals as a material for the plasma facing wall in tokamaks raised a need for in-depth studies of material mixing and the corresponding consequences on tokamak performances. Material mixing on one side influences characteristics of plasma facing components (PFC) such as its lifetime and fuel retention and on the other side the reactivity of the modified surfaces thus directly influencing edge plasma-wall interaction during discharge. Therefore detailed modelling of such complex systems requires coupling of plasma (gas) surface interaction with on-surface processes and surface-bulk material transport [1]. Binary mixing of carbon and tungsten is one of the traditional cases encountered in tokamak PFC and is also relevant to all tungsten scenario employed in ASDEX Upgrade. A study of thermal annealing of a simple double layer structure composed of evaporated carbon on clean tungsten [2] reveals the complexity of involved processes, tungsten carbide formation and its diffusion to the bulk.

We have undertaken present study in order to contribute to better understanding of hydrocarbons interaction with tungsten. Neutral hydrocarbons are abundant in the edge region of carbon-wall tokamaks and also as an impurity in all metal devices and contribute to the formation of carbon deposits. Here we present some quantitative results on H₂ formation from thermal decomposition of ethane (C₂H₆) and ethene (C₂H₄) on hot tungsten filament and filament modifications due to the carburisation under well specified experimental conditions. These results complement initial results described in [3].

Experiment

A flow cell with tungsten filament (ISPEC) was constructed as a source of vibrationally excited H₂ and D₂ molecules for special dedicated experiments. Here we used this cell to study production of hydrogen molecules through ethane and ethene interaction with hot tungsten. Cell is schematically shown in Figure 1a. It consists of two parts: i) dissociation chamber where molecules are atomized at the hot tungsten filament and ii) recombination

chamber where atom recombination occurs on the cooled wall. A removable exit aperture (EA) is mounted at the end of the cell. By simple vacuum conductivity consideration the absolute pressures can be deduced from the gas pressure measured in the vacuum chamber. The temperature of the dissociation tungsten filament is controlled by the DC current. Dissociation chamber with its hot tungsten filament has the same configuration as extensively studied reactors for Chemical Vapour Deposition (CVD) (e.g. [4]). The formation of tungsten carbide on hot filaments is also known since incandescent lamps have been in use and is especially intensively studied again in connection to CVD reactor development (e.g. [5]).

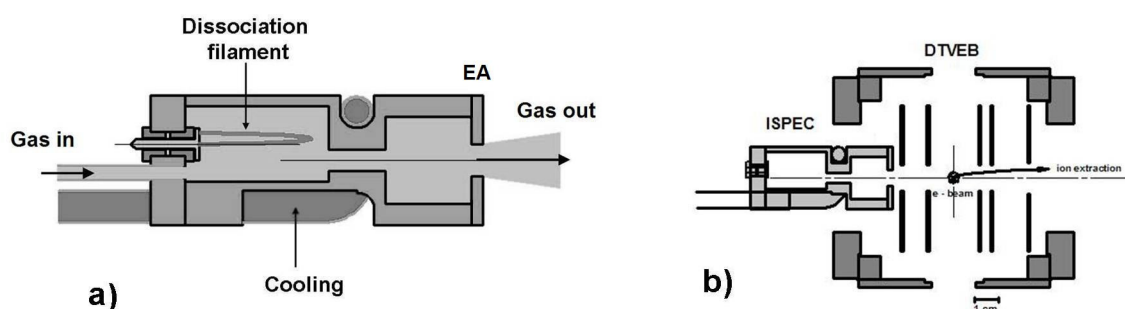


Figure 1. Experimental arrangement: a) ISPEC, and b) relative position of ISPEC mounted on vibrational spectrometer [4] DTVEB. Electron beam of the spectrometer is perpendicular to the drawing.

Recently developed vibrational spectrometer for hydrogen molecules [6] DTVEB is used in the present study for the diagnostics of the gas effusing from ISPEC. The position of ISPEC as mounted on the ion-extraction electrode system of the spectrometer is schematically shown in Figure 1b. Gas effusing from ISPEC is crossed by the electron beam of variable energy from about 0.1 to 20 eV at the distance of 25 mm. Light ions created by electron impact (negative ions from dissociative electron attachment or positive ions from non-dissociative and dissociative ionization) are extracted from the interaction region and detected by single channel electron multiplier. The spectrum of ion yield vs. electron energy is the basic diagnostic experimental information. The main and unique feature of DTVEB is that one can efficiently detect vibrationally excited H_2 (or D_2) molecules and determine their relative concentration by measuring the yield of low energy H^- (or D^-) ions from dissociative attachment of electrons in the energy range from 0 to 5 eV. Ions created below 4 eV peak originate from excited molecules and the separation of observed structures from the main peak just indicates what vibrationally excited state is contributing. From such spectra (see Figure 3) vibrational distributions of parent molecules are determined by deconvolution.

Results

When pure gas is introduced through ISPEC with cold tungsten filament then H^- (or D^-) ion yield can be detected if dissociative electron attachment leading to this fragment does

exist for the target molecule. An additional restriction for an ion to be detected ion is its energy as detection optics is tuned for the low energy ions (<200meV). Ion yield dependence on electron energy is a specific fingerprint of the target molecule because dissociative attachment is a specific resonant process. So, hydrogen has two pronounced peaks, one at 4 eV and another, about ten times bigger, at 14 eV. Detected ion yield for ethene is shown in figure 2. It appears to have contributions from three channels having maxima at 7.73, 9.06 and 10.63 eV. However it must be reminded that our detection system has strong energy selectivity so that deconvolution procedure has to be applied to obtain threshold behaviour of the cross section. H^- spectrum from C_2H_6 has different shape so that the main peak is at 9.18 eV and there is another weaker contribution peaking at 9.89 eV. In present experiments we have not observed influence of hot tungsten filament on the shape of characteristic H^- yield in both, ethane and ethene.

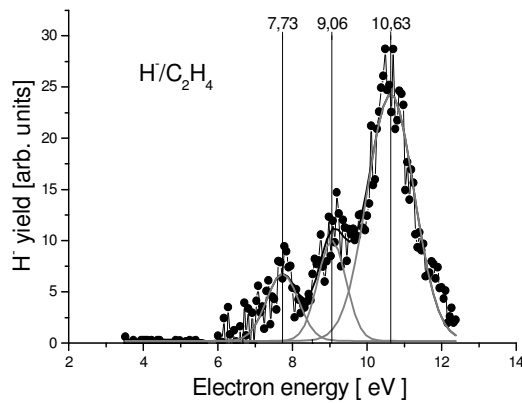


Figure 2. H^-/C_2H_4 for $I_{dis}=0A$.

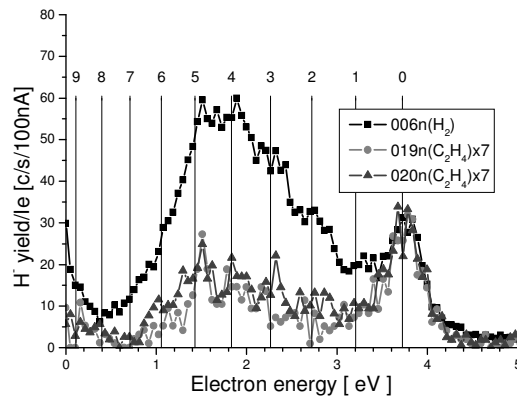


Figure 3. 4eV H^- peak from H_2 and C_2H_4 for $I_{dis}=5A$

The main result of ethane and ethene interaction with hot tungsten observed and studied in present experiment is production of hydrogen molecules by thermal decomposition of parent molecules. This is observed by appearance of two new peaks in the ion yield when filament is heated and pure ethane or ethene is in ISPEC. These two additional peaks are easily identified as originating from H_2 by their energy position and relative intensity. By comparing signal intensity for H^- from pure H_2 to H^- from H_2 originating from C_2H_4 and C_2H_6 interaction with hot tungsten one determine the absolute amount of produced hydrogen. Three spectra of 4 eV peak for H^-/H_2 obtained with filament heating current $I_{dis}=5A$ are shown in Figure 3. One spectrum is from pure hydrogen and another two are for H^- from H_2 molecules produced by ethene reaction on hot tungsten in ISPEC.

Hydrogen molecules as detected in our experiment can be produced in two distinct ways: the first is direct thermal dissociation of parent molecule at W filament leading directly to H_2 fragment and another mechanism is by surface dissociation in H-atom and then this

atom can recombine on the surface producing H_2 . From the shape of H^- spectrum at low energy side of 4 eV peak shown in Figure 3 one observes much weaker vibrational excitation for H_2 from ethene than in pure hydrogen. This indicates that in the case of ethene H_2 is produced directly on hot tungsten as secondary molecules which would be produced by atom recombination would have the same vibrational distribution as for H_2 . This second characteristics is observed in ethane thus indicating secondary molecule production.

Most of experimental conditions (pressures, temperatures, filament current and voltage) are recorded in parallel with ion yield scanning.

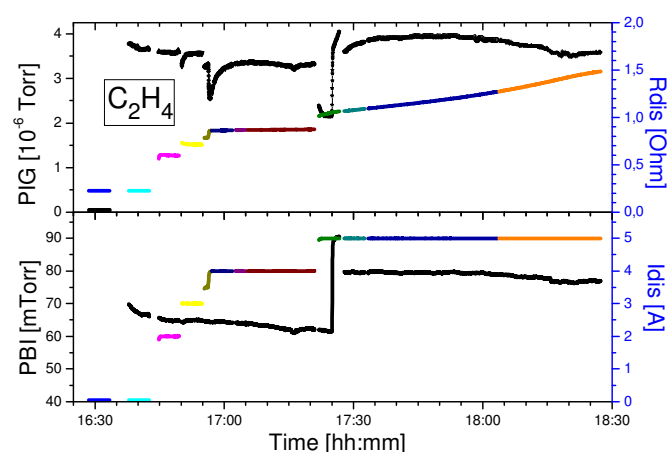


Fig. 4. Record of the pressure in the vacuum chamber and driving pressure of ISPEC and filament resistance and heating current during experiment in C_2H_4

Record of experimental conditions for the two spectra for C_2H_4 from Fig. 3 is shown in Fig. 4. A sudden but short increase of reactivity (drop of ion gauge pressure-PIG) is observed each time when I_{dis} i.e. filament temperature is increased once this temperature is above $1000^\circ C$. Corresponding increased H_2 production leads for a short time to stronger vibrational excitation. For $I_{dis}=5$ A ($1710^\circ C$) pronounced carburization of the filament starts as indicated by steady increase of the filament resistance.

References

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