

Determination of absolute atomic hydrogen densities and gas temperature by TALIF in a H₂/CH₄ microwave plasma at high pressure and high power

C. Y. Duluard*¹, X. Aubert¹, M. Wartel², C. Rond¹ and A. Gicquel¹

¹LSPM, CNRS, Université Paris 13, 99 av J.-B. Clément, 93430 Villetaneuse, France

²GREMI, Université d'Orléans, site de Bourges, Rue G. Berger, 18028 Bourges, France

*Contact e-mail: corinne.duluard@lspm.cnrs.fr

1. Introduction

This study focuses on the determination of absolute atomic hydrogen densities and gas temperature by the use of laser diagnostics, completing previous studies at lower microwave power or using Optical Emission Spectroscopy (OES) [1,2]. The TALIF technique has been implemented to determine axial profiles of H-atom densities and temperatures in a H₂/CH₄ microwave plasma.

2. Experimental set-up

The microwave (MW) reactor for diamond film deposition is a water-cooled stainless steel nearly resonant cavity operating at high pressure (100 – 250 mbar) and high MW power (2 – 3.5 kW). The discharge, produced by a 2.45 GHz MW generator, sparks off the activation of the gas mixture (0 – 4 % CH₄) leading to the formation of an approximate hemispheric plasma on the 5 cm diameter substrate holder supporting a crystal diamond substrate. The hydrogen density profile is recorded by TALIF. A dye laser pumped by a Nd:YAG laser is used for photon flux generation around 205 nm (0 – 5mJ/pulse) [3]. The laser wavelength is calibrated using an I₂ reference cell [4]. The fluorescence signal is collected with a photomultiplier tube through a 656 nm filter. The excitation beam and the collection system are mounted on motorized plates allowing measurements from 1 – 32 mm above the substrate.

3. Density calibration

The fluorescence signal intensities recorded in TALIF experiments are directly proportional to the ground state density of H atoms, however several parameters which are necessary to calculate the absolute H-atom densities cannot be determined precisely, thus TALIF measurements remain qualitative. To access absolute H-atom densities, a calibration method is needed. The method that is implemented in this study consists of normalizing the H TALIF intensity by the TALIF intensity from Kr atoms in a pure krypton gas at a known pressure. An alternative TALIF calibration method using the Kr fluorescence emission at 587.1 nm is proposed instead of the conventional calibration with the Kr

fluorescence line at 826.3 nm. In the case of TALIF measurements on H atoms, the absorption of two UV laser photons leads to the electronic excitation of 3s and 3d sublevels. In the range of H₂/CH₄ plasma pressures investigated (100 – 250 mbar), collisions with H₂ molecules, H atoms and other plasma species induce both a redistribution of the population within the n=3 sublevels (3s, 3d and 3p) and electronic quenching out of n=3 state. For calculating absolute H atom densities, a complete thermalization is assumed within the n=3 electronic state, and a global quenching rate out of the state following collisions with H₂ molecules is determined using [5]. Measurements for 1% CH₄ show that the H atom density is of the order of 10¹⁷ cm⁻³ whatever the experimental conditions.

4. Gas temperature

The gas temperature is deduced from the H_α broadening measured by TALIF. In our experimental conditions, T_e is of the order of 1 eV, thus Stark broadening is negligible, and the pressure broadening is at least one order of magnitude lower than the Doppler broadening. Several temperature measurements on atoms and molecules in similar microwave plasma reactors show that the temperature of different species is the same and is equal to the gas temperature [1]. The gas temperature is measured in H₂ and H₂/CH₄ and does not show significant difference between both mixtures. The temperature is up to 3000 K, in agreement with previous measurements [1].

References

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