

Transition phenomena and striations in inductively coupled radio-frequency plasma studied by optical emission spectroscopy

N. Čutić¹, N. Glavan^{1,2}, Z. Kregar¹, N. Krstulović¹, S. Milošević¹

¹ *Institute of Physics, Bijenička 46, HR-10000 Zagreb, Croatia*

² *Faculty of Engineering, Rijeka University, Vukovarska 58, HR-51000 Rijeka, Croatia*

Various types of standing or moving striations and transition phenomena appear in inductively coupled radio-frequency molecular (or noble gas) plasmas. When plasma is loaded with various vapours of easily evaporated liquids such as ethanol, acetone or water, the observed phenomena could be substantially modified. We present the study of such phenomena by using the optical emission spectroscopy. Inductively coupled RF plasma of hydrogen or argon is generated in the linear glass tube. Vapours are loaded into the plasma through capillary tube ending with a small orifice. The emission spectroscopy is performed longitudinally and transversally to the discharge tube using miniature fiber spectrometers. This allows a real-time monitoring of spatially resolved spectral features. Characteristic phenomena are classified in respect to the plasma parameters and the spectral appearance.

1. Introduction

Striations are well known phenomena in a various dc gas discharges. For recent review see Ref. [1]. Particular studies in radio frequency plasmas are not that frequently reported. Paired luminous rings in capacitive RF hydrogen discharges have been studied through the optical measurements [2]. Recently transition phenomena between E- to H-mode in a RF inductively coupled plasma have been studied [3]. There are numerous technological applications of inductively coupled plasmas, from surface modification [4], and surface cleaning [5-7] to sterilisation [8] where additional understanding of various phenomena is helpful in device and method development.

In the present work we have studied hydrogen and argon inductively coupled RF plasma by an optical emission spectroscopy while loading vapours of water, ethanol, methanol etc. into the plasma region.

2. Experiment

The experimental setup is shown in figure 1. The discharge vessel was a linear tube with diameter of 40 mm made of a borosilicate glass which has good transparency from 300 nm to IR. On the one side, the tube was connected to the vacuum system which consists of a one-stage oil rotary pump with the pumping speed of 35 m³/h and the base pressure of 0.01 Torr. The effective pumping speed in the discharge vessel was nearly as high as that of the pump due to appropriate large diameter vacuum connections. The pressure in the system was measured with a Baratron gauge. The flow of the gas (hydrogen, argon, etc.) was controlled by a needle

valve and measured by a mechanical flow meter. Plasma was created within a coil connected through a matching network to a RF amplifier (RIZ SW Amp.) fed by oscillation from a frequency generator (Hameg HM8131-2) with a frequency of 13.56 MHz. The power was adjustable up to 300 W and forward and reflected component were monitored at the amplifier.

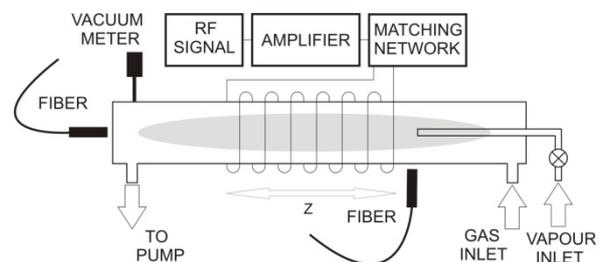


Figure 1: Experimental setup

Various liquids were placed in the small temperature-controlled glass container and introduced into the plasma region through capillary glass tube (outer diameter 0.6 cm, inner 0.1 cm). Diameter of capillary tube exit was about 0.03 cm. The amount of vapour was controlled by Teflon valve through a small hole. Prior to measurement liquids were degassed from air. The spectra were measured by means of a various optical fiber miniature spectrometers: HR2000CG-UV-NIR, HR2000-UV, LIBS2000+ from OceanOptics with nominal spectral resolution of 1 nm, 0.2 nm and 0.1 nm, respectively. They are equipped with Sony CCD arrays (2048 pixels and 12 bit A/D conversion) used as detectors. Corresponding spectral ranges were 200-1100 nm, 200-416 nm and 200-980 nm,

respectively. The spectral response was determined by means of a reference light source (LS-1-CAL, Ocean Optics for the visible-IR region and deuterium lamp for the UV spectral region). We note that special care must be taken regarding measurement of spectral response for particular integration times. Spectra were measured both longitudinally through the plasma tube and transversally at given location. The acquisition rate was ranging from 5 full spectra per second to one each few seconds. Spectrometers were controlled, and saved spectra analysed by homemade software developed within LabVIEW. Simultaneously all experiments were recorded by a standard CCD camera which was used later in the analyses.

3. Results and discussion

We present here in more detail results obtained in hydrogen and argon plasma where different striations appear. Typical features are shown in figure 2 for pure hydrogen (top) and argon (bottom) plasma. In hydrogen, pairs of luminous slices were created while in argon small plasma balls were observed.

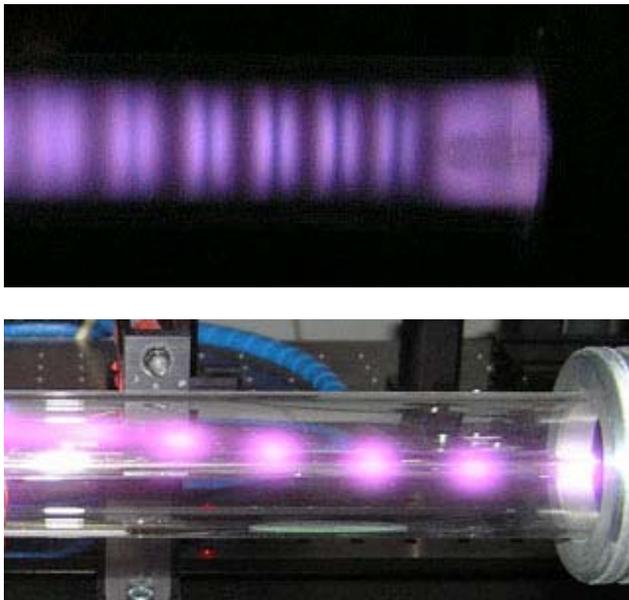


Figure 2: Luminous paired slices in hydrogen (top) and balls in argon (bottom).

These structures could be observed within a specific range of the critical pressures and discharge powers and are substantially influenced by the glass tube through which vapours are introduced into plasma (left side).

In addition, the change of the total pressure and discharge power resulted with the sudden jumps

from different plasma modes (E-mode to H-mode and vice versa).

If, for example, argon pressure was set near to the critical pressure adding water or ethanol vapour could induce various instabilities and changes of plasma modes. We performed various spectroscopic studies. In the first case, at fixed total pressure and discharge power, spatially resolved measurements along the plasma tube were performed by moving a fiber. In the second case, at fixed fiber position, temporal evolution of spectral intensities was recorded as total pressure is changed. Characteristic spectral features were identified in each case. With water vapour the OH band (309 nm), H α , β (656.2 nm 486 nm), O (777.2 nm) were observed while with ethanol, methanol, acetone the CO 3rd positive band and Angstrom band (with (0,0) transition at 283.3 nm and 450.7 nm respectively) dominates in addition to the spectral features present in a pure hydrogen or argon plasma emission.

Figure 3 shows typical time sequence of experiment where water vapour was introduced into the plasma tube. The optical fiber was placed near the capillary tube opening. Five distinct sequences were identified (A to E) as indicated in figure 3 with vertical dashed lines.

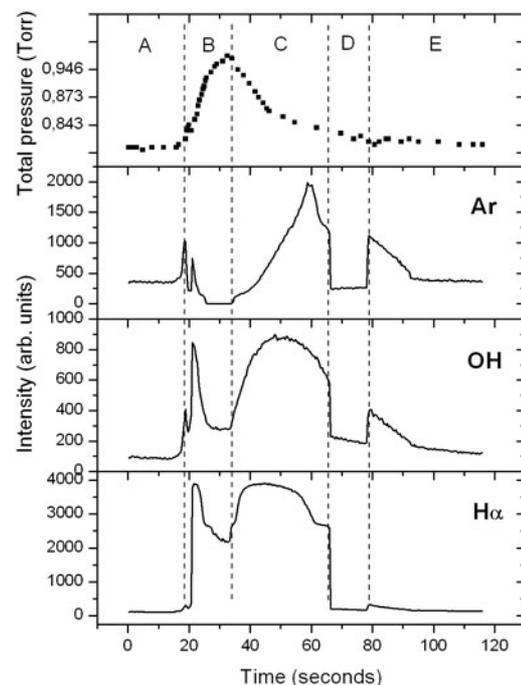


Figure 3: Time sequence of argon-water experiment.

These are: A region - pure Ar RF discharge, in stable mode, intense plasma located within a coil; B region: valve for introducing water vapour opened at the starting point, weak plasma outside a coil; C

region: water vapour valve closed at the starting point, stable plasma emission intensity slightly increasing; D region: instable plasma, moving back and forth along the coil and out; E region: stable mode, such as in region A. Figure 3 (top) shows change in total pressure with time. Starting pressure of argon was 0.836 Torr and discharge power was 130 W. Integration time of spectrometer (LIBS2000+) was 200 ms and spectra were recorded with the rate 3 per second. As water valve was opened total pressure increases by the amount determined by water vapour pressure at room temperature (23° Celsius). By closing the valve, the pressure slowly decreased towards initial value for pure argon (C region). During this period one can follow interchange of relative intensities of various spectral features (Ar at 696.635 nm, OH at 308.9 nm and H α at 656.2 nm) reflecting changes in excitation mechanisms dominating in that period of a plasma evolution. Following time region is characterised by a strong instabilities in plasma (D region) which ends as stable emission like in region A.

Figure 4 shows typical time sequence of experiment where ethanol vapour was introduced into the plasma tube. In this particular case initial parameters were such (mainly argon pressure) that a periodic oscillation exists. The B region corresponds to the ethanol valve opened and the C region corresponds the period after closing the valve. After sufficient time a periodic oscillation starts again which is related to the drop of intensities of hydrogen and CO emission.

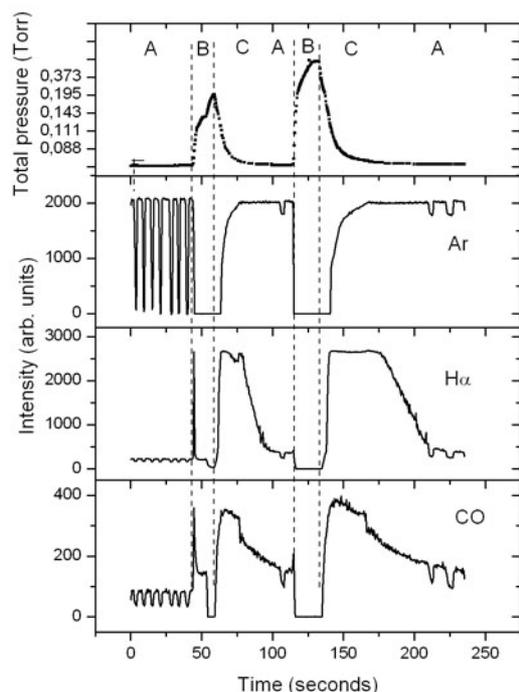


Figure 4: Time sequence of argon-ethanol experiment.

The sequence is repeated again with larger amount of ethanol vapour introduced. Comparably larger time period follows for the plasma to recover to initial condition.

Both examples shown in figure 3 and figure 4 demonstrate strong influence of vapours on electron energy distribution within plasma, as vapour molecules are decomposed and excited by electron collisions. Close inspection of molecular spectral features such as OH and CO bands shows possibility to use them for diagnostics purposes.

4. Conclusions

Striations in various RF plasmas have been studied by an optical emission spectroscopy. Fast spectra recording rate within broad spectral region allows monitoring of a plasma appearance for various plasma parameters. This approach can provide useful information for modelling of various phenomena and for applications in plasma treatment of materials.

Acknowledgement

This work has been supported by Ministry of Science Education and Sports of the Republic of Croatia under project #035-0352851-2856.

5. References

- [1] V. I. Kolobov, *J. Phys. D: Appl. Phys.* **39** (2006) R487.
- [2] Y. Sakawa, M. Hori, T. Shoji, T. Sato, *Phys. Rev. E* **60** (1999) 6007.
- [3] M. Abdel-Rahman, V. Schulz-von der Gathen, T. Gans, *J. Phys. D: Appl. Phys.* **40** (2007) 1678.
- [4] N. Krstulović, I. Labazan, S. Milošević, U. Cvelbar, A. Vesel and M. Mozetič, *J. Phys. D: Appl. Phys.* **39** (2006) 3799.
- [5] A. Vesel, M. Mozetič, A. Drenik, S. Milošević, N. Krstulović, M. Balat-Pichelin, I. Poberaj, D. Babić, *Plasma Chemistry and Plasma Processing*, Vol. 26, p. 577, 2006.
- [5] N. Glavan, N. Krstulović, N. Čutić, S. Milošević, U. Cvelbar, A. Vesel, A. Drenik and M. Mozetič, *Vakuumist*, Vol. 25, p. 29, 2005. (in Slovenian)
- [6] M. Mozetič, *Vacuum*, Vol. 61, p. 367, 2001.
- [7] M. Mozetič, *Annals of the New York Academy of Sciences*, Vol. 891, p. 325, 1999.
- [8] D. Vujošević, M. Mozetič, U. Cvelbar, N. Krstulović, S. Milošević, submitted to *J. App. Phys.* 2007.