Radiofrequency breakdown in hydrogen and deuterium: experiment and kinetic simulations

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We report the results of a combined experimental and modelling study of the breakdown of hydrogen and deuterium in radio-frequency (13.56 MHz) electric fields. The simulations are based on a simplified model to make the problem computationally tractable: only electrons are traced by Monte Carlo simulation. A very good agreement between the experimental and the calculated characteristics for hydrogen is found.

1. Introduction

The complex phenomenon of gas breakdown has been attracting considerable attention from the beginning of gas discharge research [1]. For DC excitation the details of the underlying processes are quite well understood, thanks to a large number of experimental and theoretical studies for different gases and conditions, e. g., [2-5]. In the case of RF excitation both the experiments, as well as the calculations are more complicated compared to the DC scenario [6,7].

The electrical breakdown and discharges in hydrogen have been a subject of intense research due to the wide usage of this gas in a various technological processes: thin-film deposition, etching, cleaning, etc. [8, 9]. The breakdown in hydrogen has been previously studied both for DC [10,11] and RF fields [12,13]. More recent studies of direct-current gas discharges also concentrated on micro plasma sources for which the breakdown phenomena at small electrode gaps was investigated [14].

The main aim of this work is to carry out a study of the breakdown phenomena in hydrogen and deuterium, both experimentally and by kinetic simulations based on the Monte Carlo (MC) technique [15], to provide a deeper understanding of the relevant effects.

2. Experimental

For the experiment we have built a geometrically symmetric discharge cell, which consists of two stainless steel electrodes (diameter: D = 7.5 cm) placed inside a glass cylinder at a distance of L = 1.0 cm from each other. The cell is connected to a vacuum system using 6 mm inner diameter and ~35 cm long glass tubes in order to minimize stray capacitances that could introduce an asymmetry and, thus, influence the measurements. Before the experiments, the cell is pumped down to < 10^{-6} Torr by a turbo molecular pump (TMP). We use 5.0 purity hydrogen gas ($H_2O \leq 3$ ppm, $O_2 \leq 2$ ppm, $C_nH_m \leq 0.5$ ppm, $N_2 \leq 5$ ppm) and 5.0 deuterium gas (>99.75% atom% isotopic enrichment) at a slow flow (2-6 sccm). For further purification a cryogenic trap (filled with liquid nitrogen) is used. Before starting the breakdown measurements, the electrode surface is cleaned by running (~5 mA) DC discharges with both polarities for about 10 min. For the radiofrequency measurements (f = 13.56 MHz) we have built a high-voltage push-pull oscillator similar to that described in ref. [16]. In the experiment the two electrodes are driven by the generator with potentials $U_1 = U_0 \sin(2 \pi f t)$ and $U_2 = -U_0 \sin(2 \pi f t)$, respectively. When the breakdown event occurs, the breakdown voltage is defined as the peak value of the difference of two potentials, $V_{BR} = 2U_0$. The breakdown event is detected by several methods: (a) the detection of the light of the plasma using a photodiode. (b) A change of the frequency of the high voltage oscillator (or sudden drop of the voltage amplitude) due a change of the impedance of the cell. (c) Observing an increase of the power consumption of the RF oscillator. More detailed information about the experimental set-up and the measurements methods can be found elsewhere [7].

3. Simulations

The simulation is three dimensional in velocity space and one dimensional in real space. In the simulation code only electrons are traced using Monte-Carlo technique [17], which provides a fully kinetic description under the condition of nonlocal transport. To reduce greatly the computational time, ions are not traced in the simulations. The types of collisions and the scattering processes are treated in a stochastic manner [15]. The cross sections for e- - H_2 and e- - D_2 collisions are taken from refs. [18] and [19], respectively. The initial number of electrons is 5×10^3. In the RF field, the ions, due to their mass, can respond only to time-averaged field which is zero for our conditions. Therefore, the motion of the ions in the cell can be taken as diffusive. Considering only the lowest order diffusion mode in the cylindrical geometry, the fraction of ions, f_i, that reaches the electrodes can be estimated. When an ion is created a new electron is emitted from one of the electrodes with a probability γf_i in the next RF cycle at a random time, where γ is an effective secondary electron yield. The data availability for an energy dependent electron yield for H_2^+/H^+ (or D_2^+/D^+) ions and different metals is very limited. Therefore, for our experimental conditions γ is determined from the measured DC breakdown curve using our simulation code in similar way as described in [7]. For hydrogen γ is found to be ~0.008 for low E/p. The reflection probability of electros from the electrode surface is taken to be $\rho = 0.2$. The simulation time is set to 500 RF cycles and the number of electrons in continuously monitored. The time dependent density during the last 250 cycles is then fitted by a straight line. The breakdown event is recognized when the slope of the line is near zero.

4. Results

Figure 3 illustrates the measured and simulated RF breakdown characteristics in hydrogen and deuterium at f = 13.56 MHz, the data measured in earlier studies [13] are also shown.

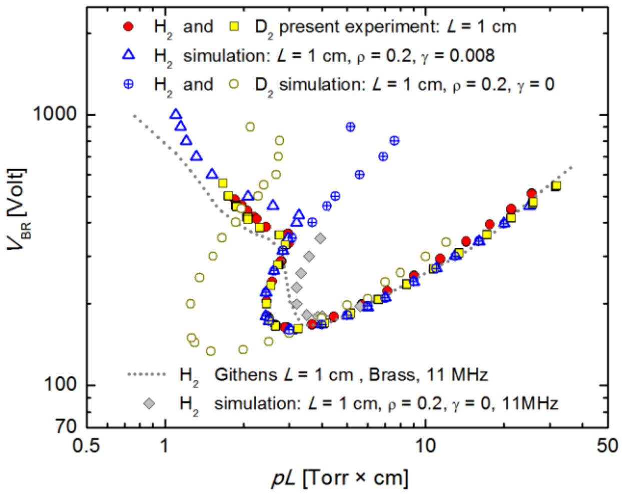

Figure 1. Comparison of the experimental breakdown voltage values in hydrogen and deuterium with simulation results. The dashed grey line represents experimental data of Githens [13] at f = 11 MHz and L =1 cm.

The shape of the measured characteristics can be explained in the following way: (i) at lower pressures, when the amplitude of the electron oscillations is comparable with the gap length, electrons can reach the surfaces and only a small fraction of them can be reflected back and create avalanches towards the opposite electrode (see Figure 2, top panel). (ii) At high pressures the amplitude of electron oscillations becomes smaller than the gap size (see Figure 2, bottom panel) and, therefore, the properties of the electrodes do not play a role. For the description of this region only gas-phase processes have to be taken into account and, thus, the accuracy of the simulation results purely depends on the accuracy and completeness of the adopted cross sections.

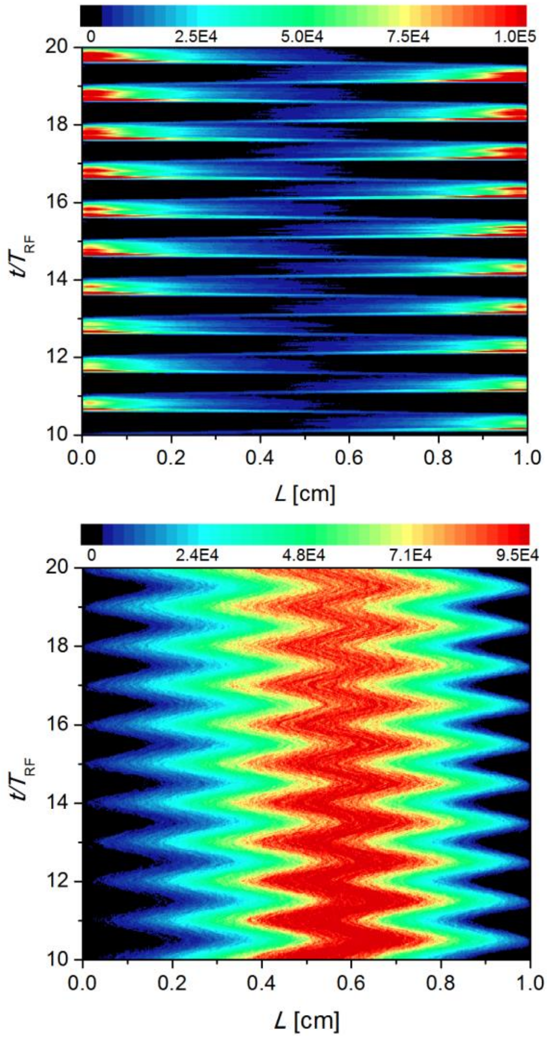

Figure 2. Spation-temporal distribution of the electron density (given in arbitrary units) during 10 RF cycles (from 10th RF cycle after starting the simulations), for points selected from the breakdown curve for hydrogen: (top) 1.2 Torr, 800 V, (bottom) 25 Torr, 460 V.

In the simulations, to reproduce the measured breakdown curve, as it was mentioned before, the secondary electron yield is taken to be γ = 0.008 - the lowest value of γ obtained for the hydrogen DC case. The results obtained from the simulation for hydrogen (see figure 1) show that, despite of the simplifications of our model, the measured curve can be accurately reproduced up to the bending point (pL = 3 Torr×cm and V_{BR} = 350 V). The slight deviation from the measured data at this point can be explained by the fact that hydrogen ions are quite light and apparently can gain some energy from the electric field and the energy dependence of γ has to be taken into account. Considering only gas-phase processes and reflection of the electrons from the electrodes we could also precisely reconstruct the experimental curve (for pL > 3 Torr×cm) measured by Githens [13] at f = 11 MHz. The horizontal shift of the 11 MHz curve toward higher pressures (with respect to the curve corresponding to 13.56 MHz) is due to the fact that at lower frequencies electrons oscillate with higher amplitude and, thus, more of them can be absorbed at the electrode surface. In contrast to the results in hydrogen, the simulated curve in deuterium shows quite a large deviation from the measured one, at the region, where the surface processes do not play role. Therefore, the deviation can be explained by the fact that the set of the cross sections used in this simulations for deuterium is not complete or incorrect, and has to be revisited.

5. Conclusions

We have carried out a combined experimental and kinetic simulation study of the breakdown in hydrogen and deuterium gases under RF (13.56 MHz) excitation. The experiments have been performed for pressures up to 30 Torr and with L = 1 cm gap length, using a glass cell, with plane parallel stainless steel electrodes.

A simulation model has been developed that traces electrons at the kinetic level. Despite the simplifications used in the model, the simulations showed that the whole breakdown curve for hydrogen can be quite accurately reproduced adopting a constant effective electron yield γ = 0.008. In case of deuterium, the deviation of the simulated breakdown curve at low pressures shows that the cross section set for e-D_2 collisions taken from ref. [20] should be carefully revisited, especially for high E/p.

Acknowledgement

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6. References

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