Two dimensional electrical modelling and experimental diagnostics of RF discharges in hydrogen

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Abstract:
A study of the electrical characteristics of RF discharges in pure molecular hydrogen at various gas pressures (ranging from 0.5 mbar to 1.5 mbar) was performed under symmetrical and asymmetrical geometries. A two dimensional PIC-MCC model coupled to external circuit equations was developed in the case of an industrial reactor. This model permits to calculate self-consistently the self-bias voltage and to study the reactor geometry effects. The corresponding calculation results (i.e. power density and self-bias voltage) are in good agreement with the experimental results.

1. Introduction
Radio-frequency (RF) discharges are usually used for thin film deposition or plasma etching or surface treatment. Furthermore, the reactor design is an important parameter governing the plasma processes because the geometry acts on both, the gas flow velocity and the gas discharge behaviours. Although the gas velocity distribution is important for the plasma process control, our focus in this paper is on the geometrical effects on the electrical discharge behaviours. Therefore, the present article is dedicated to both experimental and theoretical studies of the role of geometry on the RF discharge in molecular hydrogen.

One-dimensional (1D) simplified spherical shell fluid model has been used by Liber mann [1] in asymmetrical geometry. It is based on the classical area ratio law to study ion transport and self-bias voltage formation. In spite of the quite good estimation of the self-bias voltage, this model is not able to give the geometrical effect induced by the plasma heterogeneity due to the electric field. Since this model cannot be used, a self-consistent fluid model has been developed by Goedheer [2] using an iterative calculation of the self-bias voltage. This procedure is only able to give the steady state values. The transient phenomena require the coupling of discharge model equation with external circuit relations. Many works are then devoted to this problem [3,4,5]. For instance, Rauf and Kushner [3], have used a hybrid model. They have demonstrated that the coupling with the external circuit can strongly influence the performance of RF plasma process reactors and can allow the self-consistent calculation of the self-bias voltage.

Although in many cases 1D modelling is sufficient to give insights, 2D modelling is needed for studying geometrical effects on the electrical discharge behaviours. In the present work, a powerful 2D particle model coupled to a self-consistent calculation of the self-bias
voltage has been developed. A comparison between calculation and measurement results has been undertaken to validate our particle model.

2. Experimental set-up

Experiments were carried out in a parallel-plate RF chamber (see figure 1). The driven electrode at the bottom is connected to a 13.56 MHz RF generator by means of a matching box (comprising a blocking capacitor) while the other walls are grounded. A dielectric material surrounded the RF electrode in order to insulate it from the ground. At low power, the discharge behaved like a symmetrical discharge whereas at high power, the self-bias voltage appeared (asymmetrical situation). An additional insulator was installed between the top and the driven electrodes in order to confine the plasma between two identical electrode areas (symmetrical situation). Arbitrarily, we define the inter-electrode space in any such case as the space between the driven and showerhead electrodes (which corresponds to the symmetrical case).

The RF and dc self-bias voltages were measured using a modular probe PHV 641 recorded on a Yokogawa DL 1540CL (150 MHz) digital oscilloscope. The RF power density was determined using the subtractive method already described in reference [6].

Figure 1: Schematic lay out of the PECVD chamber with (symmetrical case) and without (asymmetrical case) additional insulator.

3. Electrical model description and Basic data

The electrical model is based on the simulation of the transport (under the electric field action) of the main charged particles involved in the RF discharge [7,8]. In the case of the particle model developed in the present work, after the selection of the dominant charged particles (electrons with positive ions), it is necessary to know the corresponding basic data, i.e., the collision cross sections of electron-molecule and ion-molecule systems for molecular hydrogen. These basic data have been taken from both experimental and theoretical data in the literature [9,10,11]. They have been fitted and completed using swarm parameter unfolding method based on Boltzmann equation for electrons and Monte Carlo method for ions. The main reactions involved in the present particle model are summarized in table 1. We have taken into account two charged particle species (electron and \( \text{H}_2^+ \) ion). In fact, the dominant ion produced by electron impact ionisation of \( \text{H}_2 \) is \( \text{H}_2^+ \) ion. It is well known that [9,10] even at low pressures, low energy \( \text{H}_2^+ \) ions are very rapidly converted into \( \text{H}_2^+ \) ones by non resonant charge exchange collisions (\( \text{H}_2^+ + \text{H}_2 \rightarrow \text{H}_2^+ + \text{H} \)). For this reason, it is realistic to consider that \( \text{H}_2^+ \) ion is the dominant ion species. Therefore, we have assumed that \( \text{H}_2^+ \) ion is formed directly by electron impact ionisation via a fictitious reaction: \( e + \text{H}_2 \rightarrow \text{H}_2^+ \) which groups reaction (12) followed instantaneously by charge exchange reaction (\( \text{H}_2^+ + \text{H}_2 \rightarrow \text{H}_2^+ + \text{H} \)).
Collision processes of electron – H$_2$ system

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Process</th>
<th>Threshold (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. e + H$_2$ → e + H$_2$</td>
<td>Elastic collision</td>
<td>-</td>
</tr>
<tr>
<td>2. e + H$_2$ → e + H$_2$ ($r$)</td>
<td>Rotational excitation</td>
<td>0.044</td>
</tr>
<tr>
<td>3. e + H$_2$ → e + H$_2$ ($r$)</td>
<td>Rotational excitation</td>
<td>0.073</td>
</tr>
<tr>
<td>4. e + H$_2$ → e + H$_2$ ($v$=1)</td>
<td>Vibrational excitation</td>
<td>0.516</td>
</tr>
<tr>
<td>5. e + H$_2$ → e + H$_2$ ($v$=2)</td>
<td>Vibrational excitation</td>
<td>1.000</td>
</tr>
<tr>
<td>6. e + H$_2$ → e + H$_2$ ($v$=3)</td>
<td>Vibrational excitation</td>
<td>1.500</td>
</tr>
<tr>
<td>7. e + H$_2$ → e + H$_2$ ($b^1\Sigma_g$) → e + 2H</td>
<td>Electronic excitation of triplet state</td>
<td>8.900</td>
</tr>
<tr>
<td>8. e + H$_2$ → e + H$_2$ ($B^1\Sigma_u$)</td>
<td>Electronic excitation</td>
<td>11.30</td>
</tr>
<tr>
<td>9. e + H$_2$ → e + H$_2$ ($c^3\Pi_u$)</td>
<td>Electronic excitation</td>
<td>11.75</td>
</tr>
<tr>
<td>10. e + H$_2$ → e + H$_2$ ($a^3\Sigma_g^+$)</td>
<td>Electronic excitation</td>
<td>11.80</td>
</tr>
<tr>
<td>11. e + H$_2$ → e + 2H</td>
<td>Dissociative excitation</td>
<td>14.70</td>
</tr>
<tr>
<td>12. e + H$_2$ → H$_2^+$ + 2e</td>
<td>Ionisation</td>
<td>15.40</td>
</tr>
</tbody>
</table>

**Collision processes of positive ion H$_2^+$ – H$_2$ system**

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Process</th>
<th>Threshold (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13. H$_2^+$ + H$_2$ → H$_2^+$ + H$_2$</td>
<td>Elastic collision</td>
<td>-</td>
</tr>
<tr>
<td>14. H$_2^+$ + H$_2$ → H$_2^+$ + H + H$_2$</td>
<td>Ion conversion</td>
<td>10.00</td>
</tr>
<tr>
<td>15. H$_2^+$ + H$_2$ → H$_2^+$ + 3H</td>
<td>Ion conversion</td>
<td>10.00</td>
</tr>
<tr>
<td>16. H$_2^+$ + H$_2$ → H$^+$ + 2H$_2$</td>
<td>Ion conversion</td>
<td>10.00</td>
</tr>
</tbody>
</table>

**Table 1:** Collision cross sections considered in our electrical model in the case of Hydrogen.

The one-dimensional model has shown the validity of these collision cross sections by comparing the calculated power density with measured values (see figure 2). Finally, a self-consistent two-dimensional Particle In Cell - Monte Carlo (PIC - MC) model has been developed. A rigorous approach is used without any restrictive hypotheses to calculate the self-bias voltage induced by the system asymmetry and the plasma spreading outside the inter-electrode space. This approach is based on coupling the RF discharge equations with boundary conditions imposed by the external circuit. Indeed, the potential at the RF electrode was calculated by taking into account on the one hand, the charged particles striking the electrode from the plasma and the charge arriving at the electrode from the external circuit and on the other hand, the induced surface charge from both sides. In addition, the solution of the classical Poisson equation ($\nabla^2 \phi = -\rho / \varepsilon$) is undertaken by decomposing, as in Vahedi et al. [12], the potential $\phi$ into a solution of Laplace equation ($\nabla^2 \phi = 0$) with boundary conditions including external circuit and a solution of a specific Poisson equation ($\nabla^2 \phi = -\rho / \varepsilon$) with zero boundary condition. This decomposition enables to reduce the computing time.

![Figure 2](image-url)
4. Results and discussions

This section concerns the results obtained by 2D symmetrical and asymmetrical models. Calculations are performed in molecular hydrogen at 260 Volts applied voltage. The inter-electrode distance is 2.7 cm and the gas temperature 393 K. The pressure is varied from 0.5 mbar to 1.5 mbar. The RF applied voltage to electrodes has the classical form at time $t$: $V_{RF}(t) = V_{max} \cos (\omega_{RF} t) + V_d$, $V_{max}$ being the maximum voltage, $V_d$ the dc self-bias voltage and $\omega_{RF}$, the RF angular velocity in rad s$^{-1}$.

It is important to notice that the measurement results, shown in figure 2, have been obtained in asymmetrical geometry at low injected power. In this injected power region, we have observed that the self-bias voltage is zero. It means that the plasma stays confined to the inter-electrode space. In order to check this hypothesis, we installed an additional thick insulator surrounding the inter-electrode space thus giving a symmetrical geometry for our reactor. In this case, we observed at first that irrespective of the injected power, the self-bias voltage is zero. Then, the measurements in asymmetrical case (without additional insulator) at low injected power are the same as in the symmetrical case (with additional insulator). These observations are also confirmed by 2D symmetrical modelling. Therefore, it is sufficient with only 1D model to describe the discharge characteristics in a symmetrical case with the advantage of a lower computing time. However, in an asymmetrical case, a 2D model is necessary to describe the discharge behaviours when the plasma spreads outside the inter-electrode space with the appearance of the self-bias voltage.

Figure 3a shows the plasma potential profile in the system without additional insulator under a pressure of 1.5 mbar. We observe that potential is positive practically in the whole gap surrounding the grounded boundaries except near the driven electrode where the potential decreases rapidly up to the value of the self-bias voltage $V_d$. This is equal to 60.7 Volts and the corresponding dissipated power, 65.51 mW/cm$^2$. The measured $V_d$, and dissipated power ($P_{d} = -62$ Volts and 65.28 mW/cm$^2$) are in quite good agreement with the above values. In figures 3b and 3c, we have presented respectively the radial ($\Omega r$) and the axial ($\Omega z$) reduced electric field in the system.

These figures show that in the vicinity of the insulator, an important electric field appears due to the high gradient of the plasma potential. This induces a strong plasma heterogeneity in this region. In fact, it has been observed that electron and ion densities are very important nearby the insulator than in the central part of the reactor where the plasma is very homogeneous.
Furthermore, the pressure dependence has been studied using the same RF voltage. Figure 4 shows the self-bias voltage as a function of the gas pressure. In fact, when the pressure increases, the self-bias voltage decreases with a good agreement between experimental and model results. Besides, we also observed that the calculations of the power density are in good accordance with the measurements. Actually, when the pressure decreases, the electron mean free path increases leading to a plasma located more at the driven electrode periphery whereas the inter-electrode space becomes a positive space charge zone. These observations are coherent with the experimental ones showing an inhomogeneous plasma deposition or treatment on the parts (about 6 cm length) of the driven electrode in the neighbourhood of the insulator.

5. Conclusion

The present work was concentrated on the geometry-effect in a plasma reactor. A 2D model was developed in symmetrical and asymmetrical configurations. After the selection of the main charged particles involved in H₂ discharge, an optimised 1D model was envisaged to estimate the dissipated power density. A good agreement was obtained with the experimental measurements. This validates at first the choice of the dominant charged particles (electron and H₂⁺ ion) and then, their collision cross sections. Besides, a 2D modelling of symmetrical discharge added a new certainty to our approach.

The 2D model in asymmetrical configuration permitted to calculate self-consistently the self-bias voltage. The good agreement between the measurement and 2D calculation concerning both self-bias voltage and dissipated power density confirmed the validity of our 2D approach involving the boundary conditions imposed by the external circuit. This modelling explained why the pressure variation could lead to a significant change in the plasma density and energy profiles and, therefore, their possible implication on the surface treatment or plasma deposition.
References